

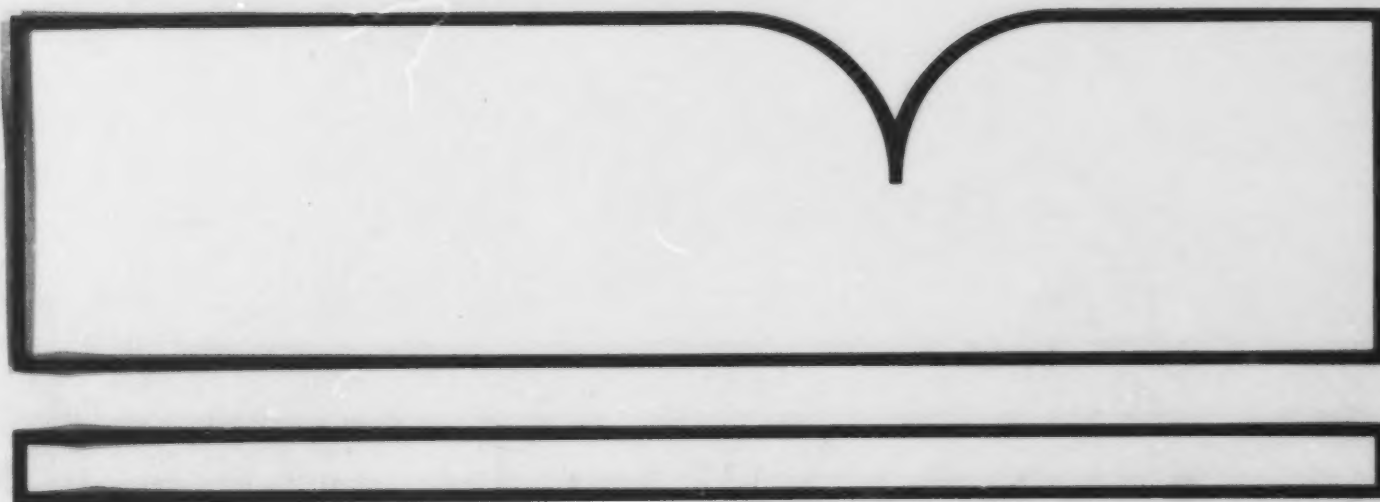
Recent Developments in SO₂ and NO_x
Abatement Technology for Stationary
Sources in Japan

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RECENT DEVELOPMENTS IN SO₂ AND NO_x ABATEMENT

TECHNOLOGY FOR STATIONARY SOURCES IN JAPAN

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FOREWORD

This report presents information on the current status and costs of air pollution control technology used in Japan. Included are reported costs of air pollution control systems installed at Japanese facilities. Caution should be used in attempting to compare the costs of U.S. control systems with Japanese control systems because the design and economic bases are different. Differences in fuel properties, regulatory requirements, cost of capital, labor, etc. make such comparisons difficult and not meaningful. For this reason, control technology costs presented in this report are in Yen. If the reader wishes to convert the reported costs to a dollar basis, he/she should do so with caution. Although the costs contained in this document are not directly related to the U.S. experience, they are useful in illustrating the relative cost differentials between different emission control concepts and between variations of a given concept.

ABSTRACT

This report is a compilation of information on the current status of abatement technology used to control major air pollutants, including SO_2 , NO_x , and particulates, in Japan. It focuses on flue gas desulfurization (FGD), combustion modification (CM), and selective catalytic reduction (SCR) of NO_x . Information presented in this report was gathered from utility company representatives and FGD, CM, and SCR process developers, as well as from the author's research in this field. Current air pollution regulations in Japan, related problems, operational parameters of commercial FGD and SCR plants, FGD and SCR economics, and the author's evaluation of the processes also are described.

TABLE OF CONTENTS

	<u>Page</u>
Foreword	iii
Abstract.....	iii
Figures.....	x
Tables.....	xv
Abbreviations and Symbols.....	xix
Acknowledgements.....	xx
Summary.....	xxi
SECTION 1	
ENERGY USE AND ENVIRONMENTAL CONTROLS.....	1-1
1.1 ENERGY SUPPLY AND SO _x /NO _x EMISSIONS.....	1-1
1.1.1 Japan's Energy Supply.....	1-1
1.1.2 Fuel Use and SO _x /NO _x Emissions.....	1-3
1.2 AIR QUALITY STANDARDS AND EMISSION CONTROL.....	1-3
1.2.1 Ambient Air Quality Standards.....	1-3
1.2.2 Emission Standards.....	1-8
1.2.3 Control Agreements.....	1-8
1.3 AMBIENT SO ₂ AND NO _x CONCENTRATIONS AND ABATEMENT STRATEGIES.....	1-11
1.4 PROBLEMS WITH AIR POLLUTION CONTROL IN JAPAN.....	1-14
1.4.1 Increase in the Number of Designated Air Pollution Patients.....	1-17
1.4.2 Increased SO ₂ Levels.....	1-17
1.4.3 Many Uneconomical FGD Plants.....	1-19
1.4.4 Ambient Sulfate Concentrations.....	1-20
1.4.5 Sulfuric Acid Mists.....	1-20
1.4.6 Photochemical Oxidants.....	1-22
1.4.7 Acid Rain.....	1-22
1.4.8 Summary.....	1-23
1.5 BEHAVIOR OF HEAVY METALS IN COAL.....	1-24
SECTION 2	
STATUS OF FLUE GAS DESULFURIZATION (FGD) IN JAPAN.....	2-1
2.1 FGD PLANTS AND BY-PRODUCTS.....	2-1
2.2 WET LIME/LIMESTONE GYPSUM PROCESS.....	2-4
2.2.1 Recent Improvements.....	2-6
2.2.2 Computer Control.....	2-10
2.2.3 Estimated FGD Cost.....	2-12
2.3 INDIRECT AND MODIFIED FGD LIME/LIMESTONE PROCESSES.....	2-12
2.4 REGENERABLE AND DRY FGD PROCESSES.....	2-17
2.4.1 Regenerable FGD Processes.....	2-17
2.4.2 Dry FGD Processes.....	2-18

TABLE OF CONTENTS (Continued)

	<u>Page</u>
2.5 CONSTRUCTION MATERIALS.....	2-19
2.6 GAS REHEATING AND WASTEWATER TREATMENT.....	2-20
2.6.1 Gas Reheating.....	2-20
2.6.2 Wastewater Treatment.....	2-22
2.7 PARTICULATE REMOVAL.....	2-24
2.8 FGD FOR COAL-FIRED BOILERS.....	2-29
SECTION 3	
NEW FGD PLANTS.....	3-1
3.1 FEATURES OF NEW FGD PLANTS FOR COAL-FIRED UTILITY BOILERS.....	3-1
3.2 EPDC'S TAKEHARA NO. 3 PLANT, (IHI).....	3-4
3.2.1 Process.....	3-4
3.2.2 Plant Flowsheet.....	3-5
3.2.3 Specifications.....	3-5
3.2.4 Performance.....	3-8
3.2.5 Economics and Plant Evaluation.....	3-8
3.3 JOBAN JOINT POWER'S NAKOSO PLANT, (MHI).....	3-8
3.3.1 Introduction.....	3-8
3.3.2 FGD Plant and Operation.....	3-10
3.3.3 Evaluation.....	3-13
3.4 NEW MITSUI MIIKE (MMC) FGD PLANTS.....	3-15
3.4.1 Development Status.....	3-15
3.4.2 Process.....	3-15
3.4.3 Performance.....	3-15
3.4.4 Economics and Evaluation.....	3-18
3.5 TOYAMA JOINT POWER'S TOYAMA PLANT (Chiyoda JBR Process).....	3-19
3.5.1 Introduction.....	3-19
3.5.2 FGD Plants.....	3-19
3.5.3 Evaluation.....	3-24
3.6 TOHOKU ELECTRIC'S SENDAI PLANT (BABCOCK HITACHI).....	3-25
3.6.1 Overview.....	3-25
3.6.2 ESP and GGH.....	3-26
3.6.3 FGD Plant.....	3-26
3.6.4 Evaluation.....	3-29

TABLE OF CONTENTS (Continued)

	<u>Page</u>
2.5 CONSTRUCTION MATERIALS.....	2-19
2.6 GAS REHEATING AND WASTEWATER TREATMENT.....	2-20
2.6.1 Gas Reheating.....	2-20
2.6.2 Wastewater Treatment.....	2-22
2.7 PARTICULATE REMOVAL.....	2-24
2.8 FGD FOR COAL-FIRED BOILERS.....	2-29
 SECTION 3	
NEW FGD PLANTS.....	3-1
3.1 FEATURES OF NEW FGD PLANTS FOR COAL-FIRED UTILITY BOILERS.....	3-1
3.2 EPDC'S TAKEHARA NO. 3 PLANT, (IHI).....	3-4
3.2.1 Process.....	3-4
3.2.2 Plant Flowsheet.....	3-5
3.2.3 Specifications.....	3-5
3.2.4 Performance.....	3-8
3.2.5 Economics and Plant Evaluation.....	3-8
3.3 JOBAN JOINT POWER'S NAKOSO PLANT, (MHI).....	3-8
3.3.1 Introduction.....	3-8
3.3.2 FGD Plant and Operation.....	3-10
3.3.3 Evaluation.....	3-13
3.4 NEW MITSUI MIIKE (MMC) FGD PLANTS.....	3-15
3.4.1 Development Status.....	3-15
3.4.2 Process.....	3-15
3.4.3 Performance.....	3-15
3.4.4 Economics and Evaluation.....	3-18
3.5 TOYAMA JOINT POWER'S TOYAMA PLANT (Chiyoda JBR Process).....	3-19
3.5.1 Introduction.....	3-19
3.5.2 FGD Plants.....	3-19
3.5.3 Evaluation.....	3-24
3.6 TOHOKU ELECTRIC'S SENDAI PLANT (BABCOCK HITACHI).....	3-25
3.6.1 Overview.....	3-25
3.6.2 ESP and GGH.....	3-26
3.6.3 FGD Plant.....	3-26
3.6.4 Evaluation.....	3-29

TABLE OF CONTENTS (Continued)

	<u>Page</u>
3.7 SHIKOKU ELECTRIC'S SAIJO PLANT (KHI Magnesium Gypsum Process).....	3-30
3.7.1 Development Status.....	3-30
3.7.2 Process Description.....	3-30
3.7.3 System Performance.....	3-35
3.7.4 Economics and Evaluation.....	3-35
3.8 NEW FGD PLANTS FOR VARIOUS GASES (Kureha, Nippon Steel, Dowa, and Chiyoda).....	3-37
3.8.1 Overview.....	3-37
3.8.2 Kureha Organic Acid-Limestone Gypsum Process.....	3-39
3.8.3 Converter Slag Scrubbing.....	3-41
3.8.4 Dowa Aluminum Sulfate-Gypsum Process.....	3-42
3.8.5 Mitsubishi Petrochemical's Yokkaichi Plant (Chiyoda CT-121 Process).....	3-45
 SECTION 4	
NO _x ABATEMENT BY COMBUSTION MODIFICATION (CM).....	4-1
4.1 NO _x ABATEMENT FOR BOILERS.....	4-1
4.2 FUNCTIONS OF CM TECHNIQUES.....	4-3
4.3 COMBUSTION MODIFICATION FOR OIL- AND GAS-FIRING.....	4-5
4.3.1 Overview.....	4-5
4.3.2 Low-NO _x Burner for Oil- and Gas-firing.....	4-6
4.3.3 Example Utility Application.....	4-8
4.4 COMBUSTION MODIFICATION FOR COAL-FIRED BOILERS.....	4-10
4.4.1 Principles of NO _x Abatement for Pulverized Coal Burning.....	4-10
4.4.2 EPDC's Tests on Low-NO _x Coal Combustion.....	4-14
4.4.3 NO _x Abatement by DF-CN _x Burner at EPDC's Isogo Station...	4-16
4.4.4 DRB and PG Dual Burner at EPDC's Takehara Station.....	4-18
4.4.5 SGR Burner and PM Burner.....	4-21
4.4.6 VD Burner.....	4-25
4.5 COMBUSTION MODIFICATION FOR VARIOUS FURNACES.....	4-25
4.5.1 Cement Kilns.....	4-25
4.5.2 Glass Melting Furnaces.....	4-27
4.5.3 Refuse Incinerators.....	4-28
 SECTION 5	
SELECTIVE CATALYTIC REDUCTION (SCR).....	5-1
5.1 OVERVIEW.....	5-1

TABLE OF CONTENTS (Continued)

	<u>Page</u>
5.2 SCR CATALYSTS.....	5-4
5.2.1 Catalyst Shapes.....	5-4
5.2.2 Catalyst Composition and Properties.....	5-4
5.2.3 Space Velocity (SV) and Area Velocity (AV).....	5-7
5.2.4 Recent Trends in SCR Catalysts.....	5-8
5.2.5 Disposal of Spent Catalyst.....	5-8
5.3 SCR PERFORMANCE.....	5-9
5.3.1 NO Removal and Unreacted NH ₃	5-9
5.3.2 Inlet NO _x Concentration and SV.....	5-11
5.3.3 Degradation of Catalyst Activity.....	5-11
5.4 SCR FOR UTILITY BOILERS.....	5-14
5.4.1 Status.....	5-14
5.4.2 Boiler Load Change and SCR.....	5-14
5.4.3 SCR Systems for Coal-fired Boilers.....	5-18
5.4.4 SCR/FGD Combination.....	5-20
5.5 SCR FOR COMBINED CYCLE POWER PLANTS.....	5-21
5.5.1 Overview.....	5-21
5.5.2 Kawasaki Plant, Japan National Railway (JNR).....	5-23
5.5.3 Higashi Niigata Plant, Tohoku Electric.....	5-25
5.5.4 Futtsu Plant, Tokyo Electric.....	5-28
5.6 SCR FOR VARIOUS GASES.....	5-30
5.6.1 Overview.....	5-30
5.6.2 Iron Ore Sintering Machine Flue Gas.....	5-31
5.6.3 Coke Oven Gas.....	5-34
5.6.4 Diesel Engine Exhaust Gas.....	5-34
5.6.5 Incinerator Flue Gas.....	5-35
5.7 SCR COST.....	5-35
 SECTION 6	
VARIOUS DRY PROCESSES FOR NO _x AND SIMULTANEOUS SO _x /NO _x ABATEMENT..	6-1
6.1 INTRODUCTION.....	6-1
6.2 SELECTIVE NONCATALYTIC REDUCTION (SNR OR THERMAL DeNO _x).....	6-3
6.2.1 Overview.....	6-3
6.2.2 Chita Plant, Chubu Electric.....	6-4
6.2.3 Thermal DeNO _x Units--Exxon Technology.....	6-4
6.2.4 SNR Units by MKK.....	6-6
6.2.5 SNR for City Refuse Incinerators.....	6-6
6.2.6 Evaluation.....	6-7

TABLE OF CONTENTS (Continued)

	<u>Page</u>
6.3 IN-FURNACE NO _x REDUCTION (IFR).....	6-7
6.3.1 Overview.....	6-7
6.3.2 Mitsubishi Heavy Industries (MHI).....	6-9
6.3.3 Hitachi Zosen's IFR Tests.....	6-11
6.3.4 Dual Combustion Denitration Process for Cement Kilns....	6-11
6.3.5 Evaluation.....	6-14
6.4 ACTIVATED CARBON (COKE) PROCESS.....	6-14
6.4.1 Introduction.....	6-14
6.4.2 EPDC-SHI Process.....	6-16
6.4.3 Mitsui-BF Process.....	6-19
6.4.4 Evaluation.....	6-22
6.5 OTHER DRY PROCESSES.....	6-22
6.5.1 Electron Beam Process.....	6-22
6.5.2 Shell Copper Oxide Process.....	6-24
6.5.3 Molecular Sieve Process.....	6-24
6.6 WET PROCESSES FOR NO _x AND SIMULTANEOUS NO _x /SO _x REMOVAL.....	6-24
6.7 FLUIDIZED-BED COMBUSTION (FBC) OF COAL.....	6-25
6.7.1 Development Status.....	6-25
6.7.2 Wakamatsu Pilot Plant (EPDC).....	6-27
6.7.3 Sakito FBC Plant (MHI-FCC process).....	6-30
6.7.4 Mitsui Aluminum Co.--Wakamatsu Plant.....	6-30
6.7.5 Evaluation.....	6-33
 SECTION 7	
ESTIMATED COSTS FOR POLLUTION CONTROL AND EVALUATION OF CONTROL METHODS.....	7-1
7.1 POWER GENERATION AND POLLUTION CONTROL COSTS.....	7-1
7.2 ACID REMOVAL COSTS.....	7-3
7.3 EVALUATION OF CONTROL METHODS.....	7-4
REFERENCES.....	R-1

FIGURES

SECTION 1

<u>Figure</u>		<u>Page</u>
1-1	Japan's primary energy supply.....	1-2
1-2	Electric power capacity in Japan.....	1-2
1-3	Annual oil and coal consumption in Japan.....	1-4
1-4	SO ₂ and NO ₂ concentrations in large cities of the world.....	1-7
1-5	Annual average ambient SO ₂ concentrations in 15 major cities and industrial regions and capacity of FGD plants.....	1-13
1-6	Annual average ambient NO ₂ concentrations in 15 major cities and industrial regions and capacity of flue gas treatment (FGT) plants for NO _x removal.....	1-15
1-7	Example flue gas treatment system for a coal-fired boiler....	1-16
1-8	Ambient SO ₂ and NO ₂ concentrations and designated air pollution patients.....	1-18
1-9	Annual average pollutant concentrations in 5 major regions in Japan.....	1-21
1-10	Number of days in a year with photochemical smog warnings (oxidants above 0.12 ppm) and average non-methane hydrocarbon concentrations at 13 automobile exhaust gas monitoring stations (based on Environmental White Paper).....	1-21

SECTION 2

2-1	Production capacity of FGD by-products.....	2-3
2-2	Price of FGD by-products.....	2-5
2-3	Flowsheet and material balance of limestone-gypsum FGD process.....	2-7
2-4	The effect of scrubber improvements on power consumption.....	2-9
2-5	L/G vs. SO ₂ removal efficiency.....	2-9
2-6	Comparison of spray tower scrubbers with varying capacities..	2-11
2-7	ASC-1,000 control system (MHI).....	2-13

FIGURES (Continued)

<u>Figure</u>		<u>Page</u>
2-8	Example of MHI's ASC-1,000 control of an FGD system and resultant power savings.....	2-14
2-9	Gas leakage in a rotary gas-gas heater.....	2-21
2-10	Typical FGD wastewater treatment system.....	2-23
2-11	Dust removal by ESP and FGD.....	2-25
2-12	Dust removal by FGD with different L/G ratios.....	2-25
2-13	Cost of dust removal by ESP and FGD.....	2-27
2-14	Particle size distribution of dust at three stages of removal.....	2-28
2-15	Wet lime/limestone processes.....	2-32
2-16	Coal blending and SO ₂ removal efficiency of the limestone-gypsum process (Babcock Hitachi).....	2-34

SECTION 3

3-1	Takehara FGD system.....	3-6
3-2	Boiler and flue gas treatment system at Joban Joint Power's Nakoso station.....	3-11
3-3	Kyushu Electric Co.-Minato FGD plant (limestone-gypsum process).....	3-17
3-4	Flowsheet for the Chiyoda CT-121 JBR Process (Toyama plant)..	3-20
3-5	Prescrubber and jet bubbling reactor (in mm).....	3-21
3-6	Submersion depth of gas sparger pipes vs. SO ₂ removal efficiency.....	3-23
3-7	pH vs. SO ₂ removal efficiency.....	3-23
3-8	Flue gas cleaning system of the No. 2 and No. 3 boilers at the Sendai station.....	3-27
3-9	Flowsheet for the Saijo plant.....	3-31
3-10	Absorbent pH, L/G ratio, and SO ₂ removal efficiency for the KHI process.....	3-32

FIGURES (Continued)

<u>Figure</u>		<u>Page</u>
3-11	Flowsheet of the Kureha organic acid limestone gypsum process.....	3-40
3-12	FGD plant using the convertor alag process at Sakai, Nippon Steel Co.....	3-43
3-13	Flowsheet of the Dowa process.....	3-44

SECTION 4

4-1	NO _x abatement methods for utility boilers.....	4-2
4-2	Effect of flue gas recirculation (FGR) and two-stage combustion.....	4-7
4-3	Four low-NO _x burner types.....	4-9
4-4	Model of low-NO _x coal burning.....	4-11
4-5	Examples of combustion modification tests conducted by EPDC and boiler manufacturers.....	4-15
4-6	IHI's boundary air system.....	4-17
4-7	Damper opening ratio and NO _x concentration (Isogo).....	4-19
4-8	Heat release rate at burner zone and NO _x concentration.....	4-20
4-9	MHI PM burner.....	4-23
4-10	Concept of pulverized coal-fired, low-NO _x PM burner.....	4-23
4-11	Comparison of NO _x emissions: laboratory test results vs. field data.....	4-24
4-12	DD process flow and operating data.....	4-26

SECTION 5

5-1	Cross section of parallel flow catalysts.....	5-6
5-2	SCR for coal-fired boiler flue gas at 350°C using a honeycomb catalyst.....	5-10
5-3	NO _x removal efficiency vs. relative amount of catalyst.....	5-12
5-4	Inlet NO _x concentration vs. relative amount of catalyst.....	5-12

FIGURES (Continued)

<u>Figure</u>		<u>Page</u>
5-5	Degradation of catalyst activity in flue gases from different fuels.....	5-13
5-6	Expected performance of an SCR unit for a utility boiler.....	5-17
5-7	Total flue gas treatment process for coal-fired boilers.....	5-19
5-8	Combined cycle plant flow diagram (JNR Kawasaki plant).....	5-24
5-9	Performance of the JNR Kawasaki Plant.....	5-26
5-10	Combined cycle power plant (Higashi Niigata Power Station, Tohoku Electric).....	5-27
5-11	Combined cycle unit (Futtsu station, Tokyo Electric).....	5-29
5-12	Systems for treatment of various flue gases.....	5-32

SECTION 6

6-1	Performance of SNR (Chita Station, Chubu Electric).....	6-5
6-2	Two-stage combustion and in-furnace NO _x reduction.....	6-8
6-3	Test of IFR with domestic low-N coal.....	6-10
6-4	Tests with imported coals.....	6-12
6-5	Tests with Austrian coal.....	6-12
6-6	Structure and function of DD furnace.....	6-13
6-7	Flow sheet of EPDC, Matsushima Station demonstration unit for dry FGD with activated coke.....	6-17
6-8	Test results of the activated carbon process at the Takehara pilot plant.....	6-18
6-9	Mitsui Mining process for removing NO _x and SO ₂ by carbon adsorption.....	6-20
6-10	Test results of electron beam process.....	6-23
6-11	NO _x abatement by two-stage combustion.....	6-28
6-12	Air ratio and combustion efficiency in MBC.....	6-28

FIGURES (Continued)

<u>Figure</u>		<u>Page</u>
6-13	Effect of two-stage combustion on SO ₂ removal.....	6-29
6-14	Effect of temperature on SO ₂ removal.....	6-29
6-15	SO ₂ removal efficiency at the Sakito FBC plant.....	6-31
6-16	NO _x concentrations at the Sakito FBC plant.....	6-31

TABLES

<u>Table</u>		<u>Page</u>
SECTION 1		
1-1	Annual NO _x Emissions from Major Stationary Sources in Japan..	1-5
1-2	Ambient Air Quality Standards in Five Selected Countries (ppm).....	1-5
1-3	NO _x Emission Standards (ppm) for Boilers in Japan.....	1-9
1-4	Recent Tightening of NO _x Emission Standards for Coal-fired Boilers (ppm, 6 percent O ₂ basis).....	1-9
1-5	Examples of Controlled NO _x Emissions from Utility Coal-fired Boilers.....	1-10
1-6	Examples of Controlled SO ₂ Emissions from Coal-fired Utility Boilers.....	1-10
1-7	Particulate Emission Standards, Emissions, and Control Technologies for Utility Boilers.....	1-12
1-8	Approximate Number of Operational Flue Gas Treating Plants in 1984.....	1-12
1-9	SO ₂ Levies in Japan from 1974 to 1984 (Yen/Nm ³ of SO ₂).....	1-19
1-10	Concentration of Elements in Coal, Oil, and the Earth's Crust (ppm).....	1-25
1-11	Estimated Emissions of Heavy Metals from 1,000 MW Coal-fired Boiler (with 200 m stack).....	1-26
1-12	Example Dissolution Test of Hazardous Components from Fly Ash (mg/l) (Environment Agency, Japan).....	1-27
1-13	Sample Fly Ash Component Dissolution Tests (mg/l) (Environment Agency, Japan).....	1-27
SECTION 2		
2-1	FGD Plants Built by Major Constructors (Operational by the end of 1984).....	2-2
2-2	Investment Cost and Power Consumption of FGD Plants for Coal-fired Boilers (Limestone-gypsum process).....	2-8

TABLES (Continued)

<u>Table</u>		<u>Page</u>
2-3	Estimated FGD Cost in 1984.....	2-15
2-4	Indirect and Modified Lime/Limestone Processes (Gypsum by-product).....	2-16
2-5	pH and Composition (mg/liter) of Wastewater from FGD Units on Coal-Fired Boilers Prior to Treatment (Environment Agency).....	2-23
2-6	FGD and SCR Units for Coal-fired Boilers (Larger than 75 MW) to be Complete by 1987.....	2-30
2-7	Classification of Wet Lime/Limestone Processes.....	2-31

SECTION 3

3-1	FGD Plants Built for Coal-fired Boilers (1979-83).....	3-2
3-2	FGD Plants for Coal-fired Boilers (1983-84).....	3-3
3-3	Major Equipment at the Takehara Plant.....	3-7
3-4	Results of Performance Tests at the Takehara FGD Plant.....	3-9
3-5	Major FGD Equipment for One Train (300 MW).....	3-12
3-6	Operation of FGD Plants at Nakoso Station.....	3-12
3-7	Comparison of Nakoso and Takehara FGD Plants.....	3-14
3-8	Operational Parameters for Two MMC FGD Plants.....	3-16
3-9	Design Basis of Toyama FGD Plants.....	3-19
3-10	Major Equipment for a Chiyoda CT-121 JBR FGD Unit.....	3-22
3-11	Regulations and Agreements for the Sendai Station Boilers....	3-26
3-12	Performance of an FGD Plant for a 175 MW Coal-fired Unit.....	3-28
3-13	FGD Specifications for Coal-fired Boilers.....	3-33
3-14	Major Equipment of the KHI FGD Plants.....	3-34

TABLES (Continued)

<u>Table</u>		<u>Page</u>
3-15	Operational Parameters of the Saijo FGD Plant.....	3-36
3-16	Major New FGD Plants for Various Gases.....	3-38
3-17	Decomposition Ratio of Organic Acids.....	3-41
SECTION 4		
4-1	Classification of CM Techniques and Their Functions.....	4-4
4-2	Combustion Modification and NO _x Abatement.....	4-5
4-3	Tokyo Electric's Oil- and Gas-fired Boilers Applying Combustion Modification (April 1981).....	4-8
4-4	NO _x Control for Oil- and Gas-fired Utility Boilers.....	4-12
4-5	Examples of Combustion Modification for Coal-fired Utility Boilers.....	4-14
4-6	Modifications Made for Increased NO _x Control at the Isogo Power Unit.....	4-18
4-7	Principal Specifications of the DD Process.....	4-27
4-8	Problems and Solutions with Two-stage Combustion for Refuse Incinerators.....	4-28
SECTION 5		
5-1	Number and Capacity of Major SCR Plants.....	5-3
5-2	Number and Capacity of SCR Units for Utility Boilers.....	5-5
5-3	Comparison of Parallel-flow Type Catalysts.....	5-5
5-4	AV and SV for a Square Honeycomb Catalyst.....	5-7
5-5	Example of NO _x Removal Conditions.....	5-9
5-6	Performance of Major SCR Plants on Utility Boilers.....	5-15
5-7	New SCR Plants on Utility Boilers.....	5-16
5-8	Comparison of High-dust and Low-dust SCR Systems for Coal-fired Boilers.....	5-18

TABLES (Continued)

<u>Table</u>	<u>Page</u>
5-9 Combined Cycle Power Plants with SCR in Japan.....	5-22
5-10 SCR for Various Gases.....	5-33
5-11 Estimated SCR Cost for New Power Plants.....	5-36

SECTION 6

6-1 Tabulation of Dry Processes for NO _x and SO _x /NO _x Abatement....	6-2
6-2 SNR Units for City Refuse Incinerators.....	6-7
6-3 Estimated Hourly Operating Cost of Mitsui-BF Activated Coke Plant for 350 MW Coal-fired Boiler.....	6-21
6-4 Oxidation/Reduction Processes and Plants.....	6-26
6-5 Complex Absorption Process.....	6-26
6-6 Coals Used for FBC Tests at Wakamatsu Pilot Plant.....	6-27
6-7 Design and FBC Operational Data of Mitsui Aluminum Wakamatsu Plant.....	6-32

SECTION 7

7-1 Coal- and Oil-fired Boiler Power Generation, Assumptions for Cost Estimation: New Utility Boilers.....	7-1
7-2 Cost of Power Generation and Pollution Control (1984).....	7-2
7-3 Acid Removal Costs.....	7-3

LIST OF ABBREVIATIONS AND SYMBOLS

APH	air preheater
AV	area velocity
BHK	Babcock Hitachi Ltd.
CM	combustion modification
EPDC	Electric Power Development Co.
ESP	electrostatic precipitator
FBC	fluidized bed combustion
FGD	flue gas desulfurization
FGR	flue gas recirculation
FGT	flue gas treatment
g	gram
GGH	gas-gas heater
h	hour
IFR	in-furnace NO _x reduction
IHI	Ishikawajima-Harima Heavy Industries
kg	kilogram
KHI	Kawasaki Heavy Industries
kl	kiloliter
km	kilometer
kW	kilowatt
l	liter
LNB	low NO _x burner
m	meter
mg	milligram
μ	micrometer = 10^{-6} m = micron
mmH ₂ O	millimeter water column
MMC	Mitsui Miike Machinery Co., now the Mitsui Miike Engineering Co.
MW	Megawatt
NM ³	normal cubic meter (0°C, 1 atmospheric pressure basis)
NO _x	nitrogen oxides
OSC	off-stoichiometric combustion
ppm	parts per million
SCR	selective catalytic reduction
SNR	selective noncatalytic reduction
SO _x	sulfur oxides
S.P.	suspended particulates
SV	space velocity
t or ton	metric ton
TSC	two-stage combustion

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SUMMARY

In order to attain stringent Japanese ambient air quality standards of 0.04 ppm (daily average) for SO_2 and 0.04-0.06 ppm (daily average) for NO_x , the emission regulations have been tightened, requiring further SO_2 and NO_x abatement. In 1984, the total number of FGD plants in Japan reached 1,400, with a total capacity of $130 \times 10^6 \text{ Nm}^3/\text{h}$ (42,000 MW equivalent). About 62% (total capacity) of the FGD plants use a wet lime/limestone process. Another 28% use sodium scrubbing. About 8% (total capacity) of the FGD plants use processes that produce sulfuric acid, elemental sulfur or ammonium sulfate by-products.

For NO_x abatement, extensive combustion modification (CM) including flue gas recirculation (FGR), staged combustion, and use of low- NO_x burners have been applied to numerous combustion facilities. The NO_x concentrations in utility boiler flue gases have been lowered to 150-300 ppm for coal-fired boilers, 80-130 ppm for oil-fired boilers, and 40-60 ppm for gas-fired boilers. For additional NO_x abatement, about 160 selective catalytic reduction (SCR) plants have been constructed. Total SCR capacity will reach $90 \times 10^6 \text{ Nm}^3/\text{h}$ (29,000 MW) equivalent by the end of 1984; of this capacity, 24,000 MW will be applied to utility boilers. About 60% of the SCR plants are used with oil-fired utility boilers, 21% are used with coal-fired boilers, and 19% for gas-fired boilers.

In 1983 and 1984, 14 coal-fired utility boilers either began operation or were scheduled to go into operation. All of these boilers use FGD to remove about 85 to 98% of SO_2 and produce a by-product gypsum. Thirteen boilers use combustion modification and SCR to remove 68-85% of NO_x . The emission concentrations produced by these 14 coal-fired boilers are 30-100 ppm SO_2 and 20-200

ppm NO_x with 20-100 mg/Nm^3 of dust. All FGD and SCR plants for coal-fired boilers have been operated with over 99% reliability.

Due to these air pollution control efforts, ambient SO_2 concentrations in Japan have been lowered to meet the stringent standard in virtually all regions of the country including large industrial areas. The ambient NO_2 concentrations have been decreasing since 1983 in spite of the rapid increase in the number of automobiles. Japan's NO_2 concentrations have met the stringent standard in most regions, even in large cities. Photochemical smog problems have been reduced as a result of the abatement of NO_x and hydrocarbons. Virtually no acid rain problems have been experienced due to effective SO_2 and NO_x abatement and Japan's topography.

The cost of both FGD and SCR has been lowered, due mainly to technology improvements. The investment cost for limestone-gypsum process FGD plants for coal-fired boilers has been lowered from about 30,000 yen/kW in 1979-1980 to about 20,000 yen/kW in 1983-1984. The power consumption of FGD processes was also reduced from 2.0-2.5% to 1.4-2.1%. The annualized cost of the wet limestone gypsum process is currently about 1.5 yen/kWh including 7 years depreciation and 10% interest.

The investment cost of SCR for utility boilers in yen/kW is 5,000-7,000 for coal-, 4,000-5,000 for oil-, and about 2,500 for gas-firing. The annualized SCR cost in yen/kWh including 7 years depreciation and 10% interest is 0.5-0.6 for coal-, 0.2-0.3 for oil-, and about 0.15 for gas-firing.

The cost of generating power by coal combustion including the cost for ash disposal, SCR, and FGD is lower than that for burning low sulfur oil, although both fuels produce a similar quality flue gas. Several large combined-cycle power plants (700-1,000 MW) that burn natural gas (either existing or planned) use SCR to remove 80-85% of NO_x .

SCR is simple and easy to operate but is much more costly than combustion modification. FGD and combustion modification may be used to prevent acid rain and SCR can be used when further NO_x abatement is needed.

Selective noncatalytic reduction (thermal DeNO_x) has been used commercially but has not become popular in Japan because of its low efficiency and high ammonia leakage. Many other NO_x removal and simultaneous SO_x/NO_x removal processes have been tested and are used commercially in small plants. Most of these processes are not used at large plants because of their high cost or by-product disposal problems.

Although Japan has succeeded in controlling SO_2 and NO_x emissions to a considerable extent, additional research is underway to improve the cost effectiveness of emission control technology.

SECTION 1

ENERGY USE AND ENVIRONMENTAL CONTROLS

1.1 ENERGY SUPPLY AND SO_x/NO_x EMISSIONS

1.1.1 Japan's Energy Supply

Japan's primary energy consumption is shown in Figure 1-1. Japan now produces about 20 million tons/year of coal and small amounts of natural gas and crude oil. These domestic fuels account for only about 6% of Japan's energy demand. In 1973, Japan had to rely on imported oil to meet 70% of its energy demand. Efforts have been made to reduce this dependency on foreign oil by increasing nuclear power and importing liquefied natural gas (LNG) and coal.

Japan imports about 70 million tons/year of coal; most of this coal is used in coke production by the steel industry. The importing of fuel coal began in 1977 and has increased rapidly since then; it is expected to reach 20 million tons in 1985 and 50 million tons in 1990.

Figure 1-2 shows the estimated electrical power generation capacity in Japan in 1983. The use of oil for power generation will decrease with the increased use of nuclear power, coal, and LNG. Between 1980-1982 when oil prices were high, many new, large coal-fired boilers were planned. However, the construction of some of these boilers has been delayed due to slow

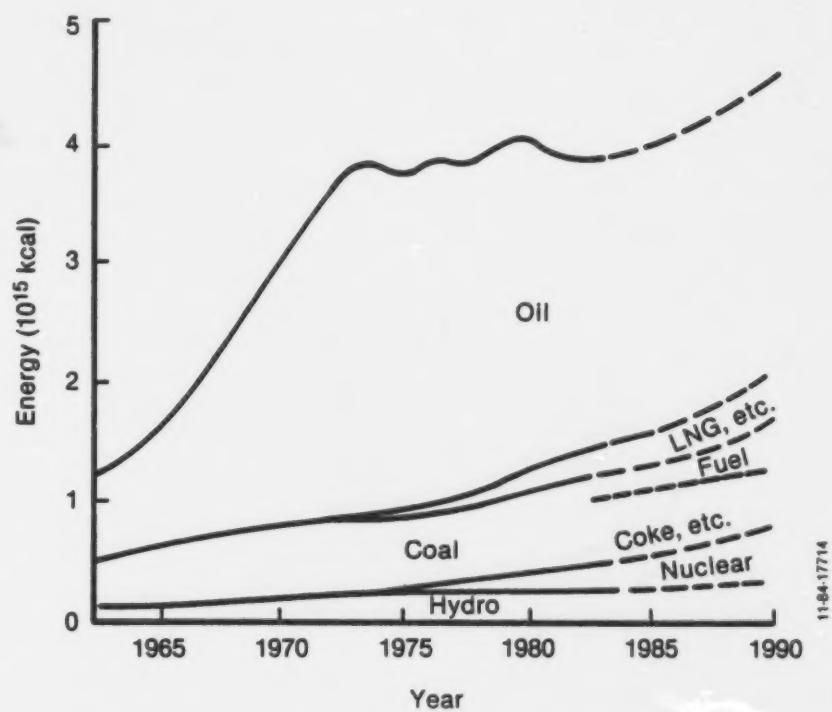


Figure 1-1. Japan's primary energy supply.

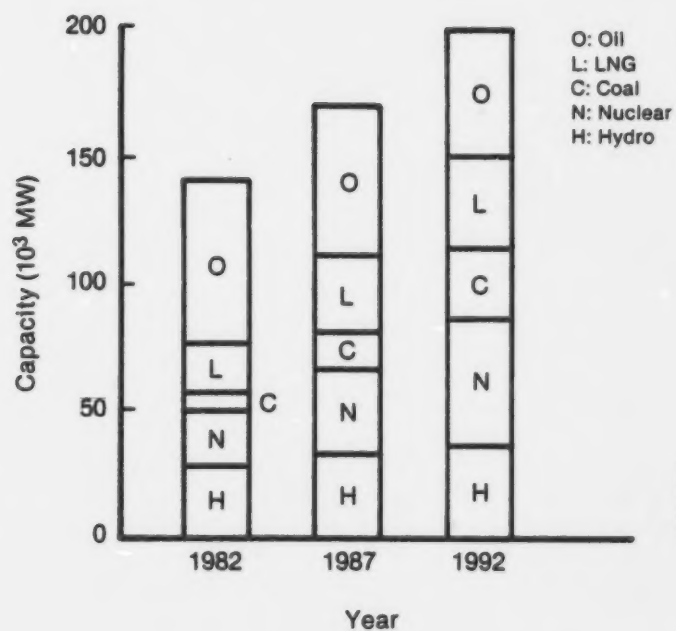


Figure 1-2. Electric power capacity in Japan.

economic growth and a recent decrease in oil prices. Power generation by coal, however, is expected to grow steadily.

1.1.2 Fuel Use and SO_x/NO_x Emissions

Since the early 1970's, most sulfur dioxide (SO_2) emissions in Japan have been produced by heavy oil combustion. The sulfur content of heavy oil has decreased, as shown in Figure 1-3, due to the importation of low-sulfur crude oil and the desulfurization of heavy oil. In addition, flue gas desulfurization (FGD) has been applied extensively to various gases. Annual SO_2 emissions in Japan have been reduced to less than about 1.5 million tons. However, the projected increase in coal consumption will necessitate further SO_2 and oxides of nitrogen (NO_x) removal in the future.

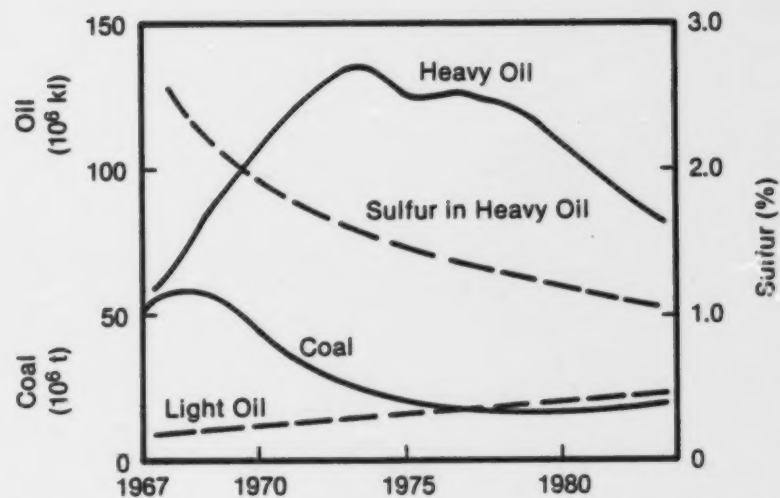
Annual NO_x emissions in Japan amount to nearly 2 million tons. In general, about 60% of these emissions are produced by stationary sources and the rest by mobile sources. However, over 60% of the NO_x emissions in large cities are produced by automobiles. Annual NO_x emissions from large stationary sources are shown in Table 1-1. Utility boilers alone emit about 300,000 tons/year of NO_x . Combustion modification (CM) and selective catalytic reduction (SCR) have been applied extensively to control NO_x emissions from stationary sources.

Several large (700-1,000 MW) LNG-fired combined-cycle power stations are either in operation or under construction. These stations have a higher thermal efficiency (41-42%) than conventional boilers (39-40%). Most combined-cycle power plants use SCR to remove 80-85 percent of NO_x emissions.

1.2 AIR QUALITY STANDARDS AND EMISSION CONTROL

1.2.1 Ambient Air Quality Standards

Ambient air quality standards for SO_2 , nitrogen dioxide (NO_2), and suspended particulates in five countries are shown in Table 1-2. As the table



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Figure 1-3. Annual oil and coal consumption in Japan.

TABLE 1-1. ANNUAL NO_x EMISSIONS FROM MAJOR STATIONARY SOURCES IN JAPAN

Source	Emissions(tons)	Emissions(%)
Utility boilers	310,000	34
Steel industry	150,000	16
Ceramic industry	130,000	14
Chemical industry	110,000	12
Oil refineries	40,000	4
Others	180,000	20
Total	920,000	100

TABLE 1-2. AMBIENT AIR QUALITY STANDARDS IN FIVE SELECTED COUNTRIES

	SO ₂ (ppm)		NO ₂ (ppm)		Suspended Particulates (mg/Nm ³)	
	Daily	Yearly	Daily	Yearly	Hourly	Daily
Japan	0.04	0.016	0.04-0.06	0.02-0.03	200	100
United States	0.14	0.03		0.05		260
West Germany		0.05		0.05		
Canada	0.06		0.10			120
Sweden	0.25					

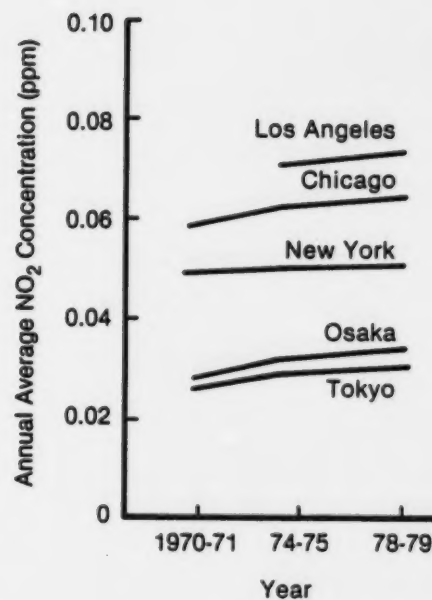
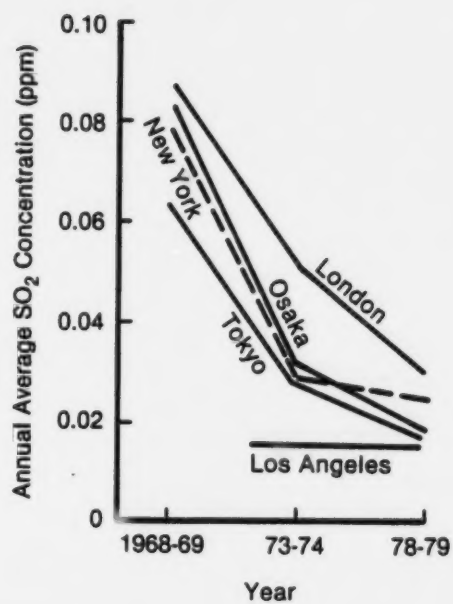
indicates, Japanese standards are more stringent than those of other countries. One reason for this is that Japanese standards originally were established as a desirable goal to keep the air clean rather than the level necessary to protect human health. Many Japanese people are afraid that they may become ill when the ambient concentrations exceed the Japanese standard and therefore want to have stringent emission regulations to maintain ambient concentrations well below the standard.

Numerous control measures have been used to attain Japan's stringent air quality standards. As a result, the ambient SO_2 concentrations have been lowered substantially, while NO_2 concentrations have been maintained at low levels. As seen in Figure 1-4, concentrations of both SO_2 and NO_2 in large Japanese cities are lower than those in most large cities in other countries. The mechanisms used to ensure attainment of air quality standards are listed below.

Emission Regulations

Emission regulations in Japan fall into the following four categories:

- (1) Emission standards established by the Central Government (Environment Agency) to restrict emissions from individual sources. These standards are applied to nearly all emission sources except private houses.
- (2) Total emission regulations established by the Central Government to control the total amount of SO_x and NO_x emissions in each region from industrial facilities with a number of emission sources. These are applied to polluted regions of Japan.
- (3) Ordinances established by prefectures and major cities and applied to large sources, including all utility power stations.



11-84-17714

Figure 1-4. SO₂ and NO₂ concentrations in large cities of the world.

- (4) Agreements between local authorities or citizen's groups with enterprises or industry. These are applied mainly to large gas sources, including utility power stations.

Generally, agreements are applied to all large gas sources and represent the most stringent type of control.

1.2.2 Emission Standards

Emission standards in Japan are based on available pollution control technology with some consideration given to economics. For example, the NO_x emission standards for stationary combustion sources are based on combustion modification (CM) technology and have been tightened (as shown in Tables 1-3 and 1-4) as the technology has been improved. NO_x emissions standards for large coal-fired boilers were 480 ppm before 1973 and 400 ppm for boilers constructed between 1973 and 1983. These standards were further tightened to 300 ppm for boilers which are built between 1983 and 1987, and to 200 ppm for those to be constructed after 1987.

Using CM, including low NO_x burners, NO_x emissions from large coal-fired boilers have been lowered to 150-200 ppm for domestic low-nitrogen coal and 200-280 ppm for imported coals. It is expected that by 1987 CM technology will be further improved so that 200 ppm may be attained for most coals. However, in-furnace NO_x reduction, a special type of CM technology, or other NO_x removal methods may be needed in order to reduce NO_x to 200 ppm with certain types of coal.

1.2.3 Control Agreements

To date, the most stringent air quality control measures have been established through local agreements as shown in Table 1-5. As a result of these agreements, selective catalytic reduction (SCR) of NO_x has been applied to most coal-fired utility boilers to reduce NO_x to 50-200 ppm. Based on control agreements, SO_2 emissions from coal-fired utility boilers have been

TABLE 1-3. NO_x EMISSION STANDARDS (ppm) FOR BOILERS IN JAPAN

Type of Boiler	Capacity (1,000 Nm ³ /h)	Year of Construction			
		pre-1973	73-75	75-77	77-83
Coal-fired (6% O ₂ basis)	Over 100	480	400	400	400
	40-100	600	480	480	400
	10-40	600	600	480	400
	Below 10	480	480	480	400
Oil-fired (4% O ₂ basis)	Over 500	180	180	150	130
	40-500	190	180	150	150
	10-40	230	230	150	150
	Below 10	250	250	250	180
Gas-fired (5% O ₂ basis)	Over 500	130	130	100	60
	100-500	130	130	100	100
	40-100	130	130	130	100
	10-40	150	150	130	130
	Below 10	150	150	150	150

TABLE 1-4. RECENT TIGHTENING OF NO_x EMISSION STANDARDS FOR COAL-FIRED BOILERS (ppm, 6 percent O₂ basis)

Capacity (1,000 Nm ³ /h)	Construction Start Year		
	pre-Sept. 1983	Sept '83 - Feb '87	Mar '87 +
Over 700	400	300	200
40-700	400	300	250
Below 40	400	300*	300*

*350 for a while.

TABLE 1-5. EXAMPLES OF CONTROLLED NO_x EMISSIONS FROM UTILITY COAL-FIRED BOILERS

Power Company	Station	New or Retrofit	Capacity (MW)	NO _x , ppm		Actual Emissions
				Emission Standard	Local Agreement	
EPDC ^a	Isogo ^b	R	265	400	159 ^c	150-180 ^d
EPDC	Matsushima ^e	N	500	400	300	230-270 ^d
EPDC	Takehara	N	700	400	-	50-60 ^f
Chugoku Electric	Shimonoseki	R	175	400	200	180-190 ^f
Hokkaido Electric	Tomato-Atsuma	N	350	400	170	150-160 ^f
Tohoku Electric	Sendai	R	156	480	260	180-200 ^f

^aElectric Power Development Co.^bLocated in urban area^cRequested by local government; the figure is under negotiation^dAttained by combustion modification^eLocated in rural area^fBy combustion modification and selective catalytic reductionTABLE 1-6. EXAMPLES OF CONTROLLED SO₂ EMISSIONS FROM COAL-FIRED UTILITY BOILERS

Power Company	Station	New or Retrofit	Capacity (MW)	SO ₂ , ppm		Removal (%)
				FGD in	FGD out	
EPDC ^a	Isogo ^b	R	265	450	20	95.5
EPDC	Takasago	R	250	1,500	130	91.3
EPDC	Takehara	R	250	1,700	100	94.1
EPDC	Takehara	N	700	700	35	95.0
EPDC	Matsushima ^c	N	500	1,000	90	91.0
Chugoku Electric	Shimonoseki	R	175	730	45	93.8
Hokkaido Electric	Tomato-Atsuma ^d	N	350	230	20	91.3
Tohoku Electric	Sendai	R	175	360	27	92.5

^aElectric Power Development Co.^bIn a large city^cIn a rural area. Three-fourths of the gas is subjected to FGD^dHalf of the gas is subjected to FGD

lowered to 20-130 ppm through the use of flue gas desulfurization (FGD) (Table 1-6).

Stringent regulations in Japan have promoted the development of emissions control technology. For example, the most stringent NO_x emission standard for automobile exhausts--0.25g/km for model year 1978 and newer gasoline-engine passenger cars--has resulted in a marked improvement in engines. These engines are able to meet the standard and also consume less gasoline.

Table 1-7 shows regulations, emissions, and particulate removal processes for utility boilers. Compared to regulations for SO_2 and NO_x emissions, particulate regulations are relatively lenient. The particulate emission standard for new coal-fired boilers in the U.S. is 30 mg/Nm^3 , much more stringent than the comparable emission standard in Japan. Ambient particulate concentrations have been lowered but still exceed the air quality standards in large cities and industrial regions in Japan. Recently the emission standards have been tightened. For new large boilers, the new standards (in mg/Nm^3) are 50 for coal-, 40 for oil-, and 30 for gas-firing in polluted regions and 100 for coal- and 50 for oil- and gas-firing in general regions.

1.3 AMBIENT SO_2 AND NO_x CONCENTRATIONS AND ABATEMENT STRATEGIES

As shown in Figure 1-5, ambient SO_2 concentrations in Japan were high in the late 1960s in large cities and industrial regions. Between the late 1960s and the early 1970s, SO_2 was reduced mainly through the importation of low-sulfur fuels and the desulfurization of heavy oil. This resulted in a decrease in ambient SO_2 concentrations in polluted regions; in 1972, the annual average ambient SO_2 concentration was 0.03 ppm. FGD capacity has increased rapidly since 1972, resulting in the reduction of annual average ambient SO_2 concentrations to 0.02 ppm. This level is sufficient to meet the stringent Japanese standard in virtually all regions of the country. There are now 1,400 FGD plants with a total capacity of 42,000 MW (Table 1-8).

TABLE 1-7. PARTICULATE EMISSION STANDARDS, EMISSIONS, AND CONTROL TECHNOLOGIES FOR UTILITY BOILERS

Power Company	Station	Fuel	Capacity (MW)	Emission Standard	Agreement mg/Nm ³	Actual Emissions	Control Technologies in Use
EPDC	Takehara	Coal	250	200	40	40	ESP+FGD
EPDC	Takehara	Coal	700	200	30	25	ESP+FGD
EPDC	Matsushima	Coal	500	200	100	70	ESP+FGD
Hokkaido	Tomato	Coal	350	200	34	30	ESP+FGD
Chugoku	Shimonoseki	Oil	400	100	40	30	FGD+ESP
EPDC	Takehara	Oil	350	100	20	20	ESP

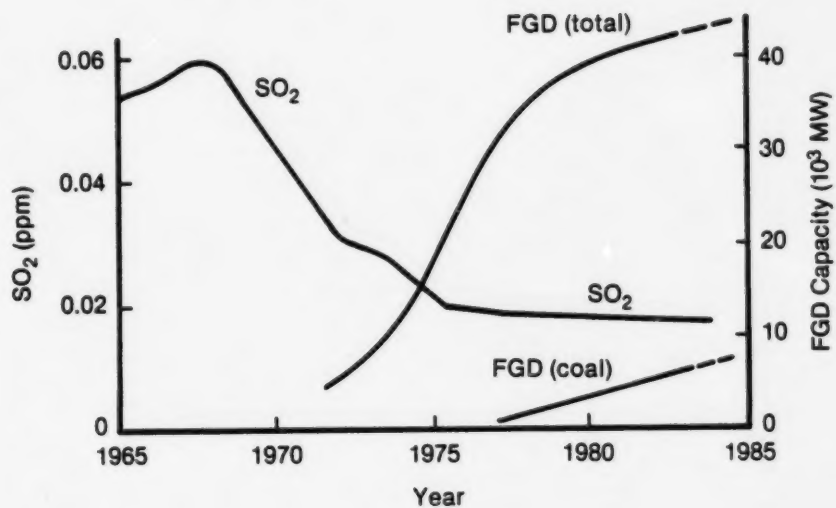
TABLE 1-8. APPROXIMATE NUMBER OF OPERATIONAL FLUE GAS TREATING PLANTS IN 1984

	Japan	United States	West Germany	Others
FGD*	1,400	250	10	10
SCR**	160	1***	0***	0***

*Flue gas desulfurization.

**Selective catalytic reduction of NO_x.

***There may be a few SCR plants for waste gases from nitric acid and nitrophosphate plants.



11-84-17714

Figure 1-5. Annual average ambient SO₂ concentrations in 15 major cities and industrial regions and capacity of FGD plants.

For NO_x abatement, combustion modification, including flue gas recirculation, staged combustion, use of low- NO_x burners, and fuel switching have been applied to numerous gas sources. In addition, about 200 plants which utilize flue gas treatment for NO_x removal have been constructed. These plants have a total capacity of about 30,000 MW equivalent.

Selective catalytic reduction technology has been improved and now is used in 160 plants which account for about 95 percent of the total capacity of the NO_x removal plants (Figure 1-6). Most coal-fired boilers use both SCR and FGD (Figure 1-7). SCR capacity has increased rapidly since 1978 as utility power companies have begun to apply SCR to both new and existing boilers.

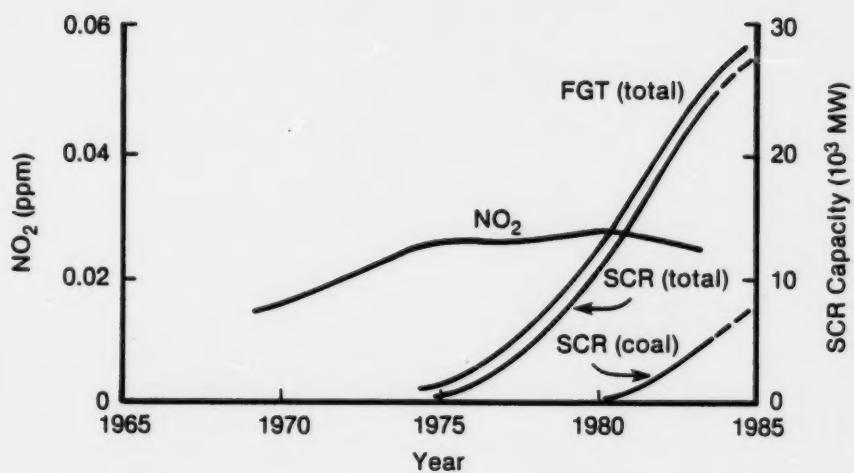
There are about 40 NO_x removal plants using processes other than SCR, such as selective noncatalytic reduction (Thermal DeNO_x) and wet processes. However, application of these processes has been limited, as will be described in Section 6 of this report.

Since 1978, NO_x emissions from gasoline-engine passenger cars have been strictly limited to 0.25 g/km. Diesel-engine automobiles also have been controlled by increasingly stringent regulations.

Through these efforts, the ambient NO_2 concentrations in Japan have been decreasing since 1980 in spite of a rapid increase in the number of automobiles. Ambient SO_2 and NO_2 concentrations in large cities and industrial regions in Japan generally are lower than those in other countries.

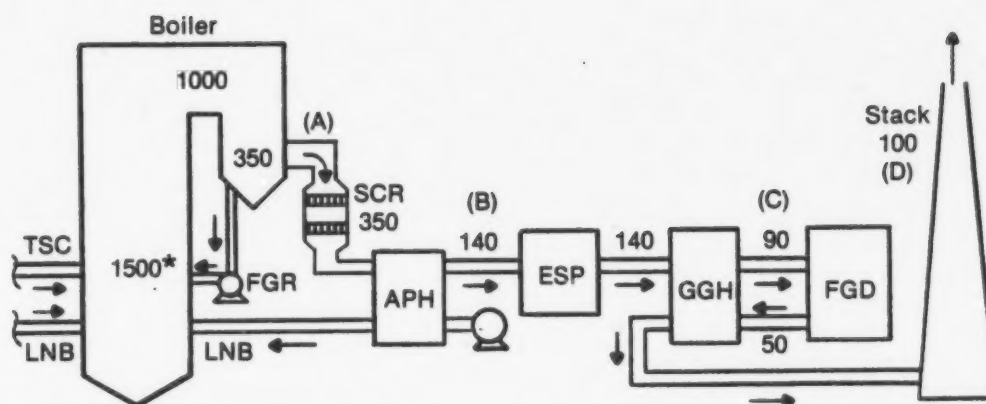
1.4 PROBLEMS WITH AIR POLLUTION CONTROL IN JAPAN

Through the use of air pollution control, Japanese air quality has improved even in large cities and industrial regions. Photochemical smog problems have been considerably reduced and virtually no acid rain problem exists. Despite this improvement, however, Japan is experiencing continuing air pollution control problems, as discussed in the following subsections:



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Figure 1-6. Annual average ambient NO_2 concentrations in 15 major cities and industrial regions and capacity of flue gas treatment (FGT) plants for NO_x removal.



*Numbers Indicate Gas Temperature, °C

	(A)	(B)	(C)	(D)
Dust (g/Nm ³)	20	20	0.1	0.02
SO ₂ (ppm)	1,000	1,000	1,000	100
NO _x (ppm)	300	90	90	90

TSC: Two-stage combustion
 LNB: Low-NO_x burner
 APH: Air preheater
 GGH: Gas-gas heater

FGR: Flue gas recirculation
 SCR: Selective catalytic reduction
 ESP: Electrostatic precipitator
 FGD: Flue gas desulfurization

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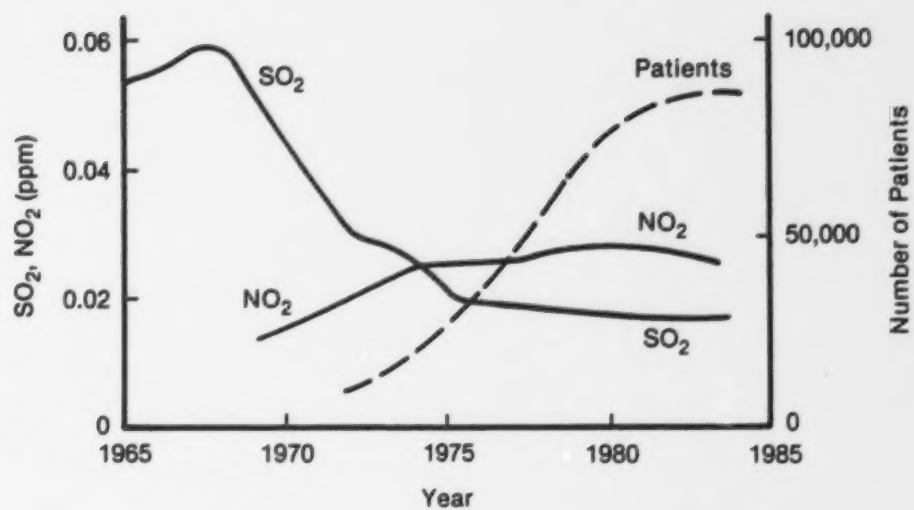
Figure 1-7. Example flue gas treatment system for a coal-fired boiler.

1.4.1 Increase in the Number of Designated Air Pollution Patients

The number of designated air pollution patients in Japan has increased substantially, as shown in Figure 1-8, while ambient SO_2 concentrations have decreased to a very low level. Designated patients are individuals who live in designated SO_2 pollution regions of the country and who are diagnosed as being air pollution victims. The rapid increase in the number of these patients between 1976 and 1980 was due mainly to the expansion of the designated pollution regions to include most sections of such large cities as Tokyo and Osaka. The number of designated patients continues to rise and some groups claim that NO_2 emissions are partially to blame. Ambient NO_2 concentrations, however, are well below the U. S. standard of 0.05 ppm except in a few areas with heavily traveled roads.

1.4.2 Increased SO_2 Levies

Since 1974, levies have been applied to SO_2 sources that emit more than 5,000 Nm^3/h (1.5 MW equivalent) of exhaust gas. Owners of SO_2 sources must pay a levy according to the amount of SO_2 emitted, even if their source is complying with SO_2 regulations. The money collected through this levy is used to aid the designated air pollution patients and exceeds 50 billion yen/year. The levy rate has increased substantially in the last ten years (Table 1-9), because the number of designated patients has increased drastically even though SO_2 emissions have been reduced. In many of the designated pollution regions, both SO_2 and NO_2 concentrations have been lowered to meet stringent air quality standards. The Federation of Economic Organizations wants to remove these clean areas from the list of designated regions to reduce the overall number of designated patients. On the other hand, some groups claim that the patients' health problems are caused by NO_x and want to establish an NO_x levy.



11-84-17714

Figure 1-8. Ambient SO_2 and NO_2 concentrations and designated air pollution patients.

TABLE 1-9. SO₂ LEVIES IN JAPAN FROM 1974 to 1984 (Yen/Nm³ of SO₂)

Regions	1974	1976	1977	1979	1981	1983	1984
Osaka	16	210	536	1239	1564	2559	3063
Tokyo	16	210	381	646	923	1548	1854
Chiba, etc.	16	210	345	517	722	1346	1612
Omura, etc.	16	210	307	452	522	942	1128
General	2	23	34	72	89	149	179

1.4.3 Many Uneconomical FGD Plants

The SO₂ levy program has promoted the construction of FGD plants, including about 1,000 small plants with a unit capacity of 3,000-30,000 Nm³/h (1-10 MW). These small plants are not very cost-effective and often are operated without sufficient control. To reduce their levies, owners of SO₂ sources tend to construct new FGD plants or to increase the SO₂ removal efficiency of existing FGD plants, even though their emissions already are low enough to meet the regulations. As will be detailed further in Section 7 of this report, SO₂ may be removed at a cost of 7-17 yen/mole or 310-750 yen/Nm³, and the cost of FGD is lower than the levy in many regions (Table 1-9).

It is strange that both SO₂ and NO₂ concentrations have been lowered in nearly all regions to meet the stringent regulations and yet the designated patients are increasing. This situation may have to be studied in order to determine the real reason for the increase in patients and more efficient methods of pollution control.

1.4.4 Ambient Sulfate Concentrations

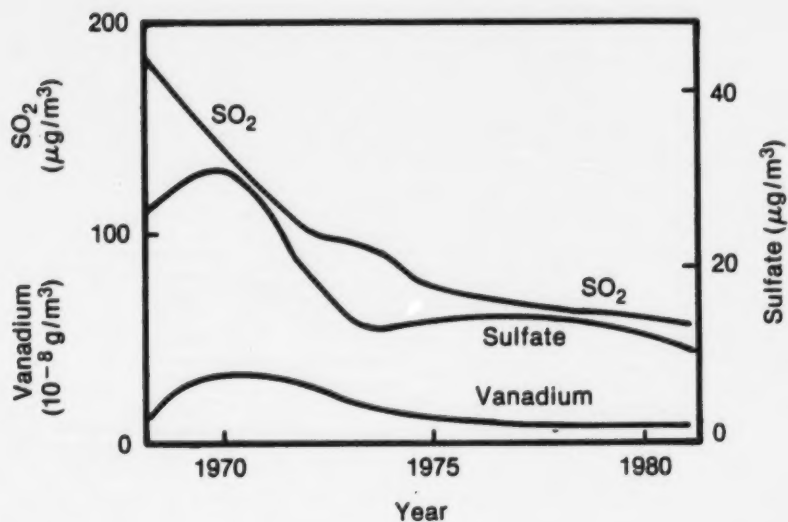
Sulfates, particularly sulfuric acid mists, may have a greater impact than SO_2 on human health. Annual average sulfate concentrations in five large Japanese cities and industrial regions--Tokyo, Osaka, Kawasaki, Amagasaki, and Kita-Kyushu--are shown in Figure 1-9. In the late 1960s, ambient SO_2 concentrations decreased, but sulfate increased along with an increase in vanadium. This may be due to fuel switching from coal to oil with a lower sulfur content. Oil-burning usually produces a much larger amount of SO_3 and vanadium than does coal. This phenomenon was also observed in the U.S. in the 1960s (1).

In Japan, ambient sulfate and vanadium both decreased between 1970 and 1973, presumably because many ESPs and FGD plants were constructed during that time. FGD usually removes about 90% of SO_2 , 50% of SO_3 , and 60-90% of vanadium. The slight increase in ambient sulfate between 1973 and 1977 may be due to high oxidant concentrations (Figure 1-10) which promote oxidation of SO_2 , and to an increase in diesel-engine automobiles using light oil containing 0.4 percent sulfur.

The decrease in sulfate concentrations since 1978 may be a result of the following: 1) a decrease in oxidants; 2) application of extensive combustion modification for NO_x control which also reduces the formation of SO_3 ; 3) improvements in mist eliminators at FGD plants, including the use of wet ESPs to remove over 95 percent of sulfates; and 4) the 1980 modification of the analytical procedure used to measure sulfate which gives a slightly smaller value of sulfate.

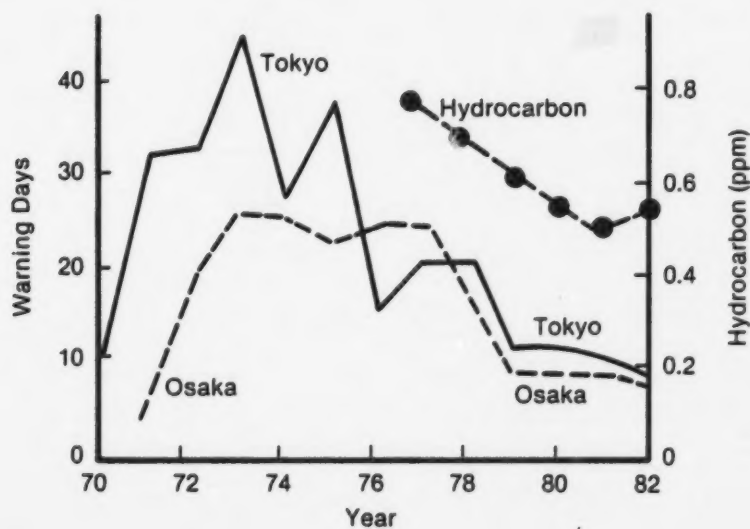
1.4.5 Sulfuric Acid Mists

Sulfuric acid mists may be divided into two types: 1) mists of concentrated sulfuric acid (over 50 percent H_2SO_4) derived from SO_3 in waste gases, and 2) mists of dilute sulfuric acid (below 0.1 percent H_2SO_4) derived from SO_2 which is slowly oxidized in the air in the presence of moisture. The



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Figure 1-9. Annual average pollutant concentrations in 5 major regions in Japan.



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Figure 1-10. Number of days in a year with photochemical smog warnings (oxidants above 0.12 ppm) and average non-methane hydrocarbon concentrations at 13 automobile exhaust gas monitoring stations

first type of mist exists in smaller quantities than the second, but has a greater impact on human health. The second type of mist is more closely associated with the formation of acid rain. A recent study of the cause of the Yokkaichi Asthma, which was the original reason for stringent SO_2 control in Japan, showed that asthma was caused not by SO_2 but by mists of concentrated sulfuric acid (2).

Although ambient sulfate concentrations in Japan are not high, the concentrated sulfuric acid mists derived from SO_3 in vehicle exhaust gas may have an impact on the health of individuals in crowded urban areas. It does not appear to make sense for power stations and industrial plants in and near large cities to be required to control emissions by burning low-sulfur (less than 0.2%) oil or natural gas or using tall stacks, while light oil containing 0.4% sulfur is burned on the streets. In the U.S., light oil generally contains about 0.2% sulfur while power stations burn fuels with higher sulfur content. The sulfur content of light oil in Japan could be reduced. Japan already has many plants capable of reducing sulfur in heavy oil to as low as 0.1% and it is easier to desulfurize light oil than heavy oil.

1.4.6 Photochemical Oxidants

As shown in Figure 1-10, photochemical oxidant concentrations in large Japanese cities were high in 1973-1977, causing photochemical smog problems. Many people strongly urged that NO_x be reduced since NO_x was considered to be the main cause of the oxidants. Oxidant concentrations have been reduced substantially since 1977 while NO_x levels increased until 1980. It appears that the reduction of oxidants has been due mainly to the reduction of non-methane hydrocarbons (Figure 1-10).

1.4.7 Acid Rain

Acid rain problems are less significant in Japan than in North America and Europe. There has been virtually no damage to trees and fish; the major

problem has been eye irritation. In the early summer of 1974 and 1975, more than 10,000 people suffered from eye irritation caused by the fine rain drops that fall at the beginning of a rain shower. Although the overall pH of the rain was 3.5, tests showed that some of the rain droplets at the beginning of the rainfall had a pH as low as 2.5. The author's tests on his own eyes revealed that any acid with a pH of 2.5 or below irritates eyes. Therefore, eye irritation may be caused by rain droplets with the pH of 2.5 or below. Widespread eye irritation was reported in 1974 and 1975 when ambient oxidants were high, especially in the early summer months. However, no eye irritation was reported before 1973 when the annual average SO_2 and sulfate concentrations were higher than in 1974-75, nor since 1977. It appears that acid rain was caused by high oxidant concentrations which promoted the oxidation of SO_2 to sulfuric acid and also by special weather conditions conducive to SO_2 oxidation: little wind, and high humidity and temperature.

The lack of a serious acid rain problem in Japan is due to SO_2 and NO_x abatement and the topography of the country. Annual SO_2 emissions in Japan currently are less than 2 million tons. By comparison, SO_2 emissions are about 3 million tons in West Germany with a land space 60 percent as big as Japan. Moreover, Japan is surrounded by ocean while West Germany is surrounded by industrial countries that emit SO_2 . The predominant wind in Japan is from west to east; it disperses flue gases out over the Pacific Ocean. In the future, Japan may suffer from acid rain caused by SO_2 and NO_x emitted by Korea and China where industry is growing rapidly without SO_2 and NO_x control. Japan is about 200-1,000 km downwind of Korea and China, within a suitable range to be affected by acid rain.

1.4.8 Summary

As the result of air pollution control efforts, haze and photochemical smog around large cities and industrial regions in Japan have been reduced considerably. This reduction, however, may be due more to the control of particulates and hydrocarbons rather than SO_2 and NO_x abatement. Japan is made up of small narrow islands; there is no land downstream of the

predominant wind. Thus SO_2 and NO_x abatement may be less important here than for countries on the continent.

1.5 BEHAVIOR OF HEAVY METALS IN COAL

Table 1-10 shows concentrations of minor elements found in coal compared with those in oil and the earth's crust. The elements present in coal in concentrations more than ten times larger than in the earth's crust are boron (B), cadmium (Cd), and selenium (Se). B and Se, as well as arsenic (As), copper (Cu), manganese (Mn), molybdenum (Mo), zinc (Zn), etc., are essential for plant and animal growth, but can be hazardous in high concentrations.

Table 1-11 shows the estimated emissions of heavy metals produced by pulverized coal combustion. In Japan, a combination of an electrostatic precipitator (ESP) and wet FGD is used to remove 99.7-99.9% of the fly ash in this type of flue gas. Many heavy metals and their compounds are volatile at a high combustion temperature and may not be as efficiently removed by a cleaning system as fly ash. Table 1-11 assumes that 50% of the mercury and 95% of the other heavy metals have been removed. Yet the ground level concentrations of heavy metals are far below the safety level. Accumulation of heavy metals in soil also may not produce any adverse effect because those metals are contained in natural soils in concentrations nearly equal to those in coal (Table 1-10).

Most of the heavy metals produced by pulverized coal combustion goes into fly ash. This fly ash may not be hazardous when used as landfill or dumped in the ocean because few heavy metals are water soluble (Tables 1-12 and 1-13). In Japan, a considerable amount of fly ash is used as a soil conditioner to supply micronutrients such as boron, zinc, and manganese to plants.

Heavy metals in coal fly ash do not appear to produce any significant adverse effects on environment as long as the fly ash is caught efficiently and used or disposed of properly. However, further studies may be needed to learn more about the behavior of submicron particles from coal which are not

TABLE 1-10. CONCENTRATION OF ELEMENTS IN COAL, OIL, AND THE EARTH'S CRUST (ppm)

Element	Coal ^a			Oil ^b (ppm)	Earth Crust (B) ^c	A/B
	Average (A)	Min.	Max.			
As	14.02	0.53	93.00	0.26	2	7
B	102.21	5.00	224.00	0.002	3	34
Be	1.61	0.20	4.00	<0.003	2	1
Cd	2.52	0.10	65.00	0.03	0.15	17
Co	9.57	1.00	43.00	0.05	<20	1
Cr	13.75	4.00	54.00	0.05	100	0.1
Cu	15.16	5.00	61.00	1.3	55	0.3
F	60.94	25.00	143.00	—	700	0.1
Hg	0.20	0.02	1.60	0.05	0.08	2.5
Mn	49.4	6.00	181.00	0.6	1,000	0.05
Mo	7.54	1.00	30.00	80.00	10	0.7
Ni	21.07	3.00	80.00	10.00	<35	1
Pb	34.78	4.00	218.00	—	15	2
Sb	1.26	0.20	8.90	0.05	0.1-0.1	10
Se	2.08	0.45	7.70	0.03	0.09	23
V	32.71	11.00	78.00	50	120	0.3
Zn	272	6.00	5,350	30	40	6.3
U	1.6	0.5	4.5	0.015	3-4	0.5
Th	2.01	1.2	3.3	—	10-15	0.1
Cl	1,400	100	5,400	39	150	9.3

^aN.Y. Lim, "Trace Elements from Coal Combustion Atmospheric Emissions," IEA-Coal Research, London ICTIS/TRO 5 (1979)

^bEnvironmental Chemistry (1979)

^cAhrens-Taylor (1961), etc.

TABLE 1-11. ESTIMATED EMISSIONS OF HEAVY METALS FROM 1000 MW COAL-FIRED BOILER (with 200 m Stack)

Metals	Content in Coal (ppm)	In Combustion Gas ($\mu\text{g}/\text{Nm}^3$)		In the Atmosphere ($\mu\text{g}/\text{Nm}^3$)	
		Before Cleaning ^a	After Cleaning ^c	Ground Level	Permissible Concentration ^c U.S.S.R. ^d Standard
Mercury (Hg)	.1-1	10-100	5-50	10^{-4} - 10^{-3}	50 0.3
Cadmium (Cd)	1-10	10^2 - 10^3	5-50	10^{-4} - 10^{-3}	50 --
Beryllium (Be)	1-10	10^2 - 10^3	5-50	10^{-4} - 10^{-3}	2 --
Selenium (Se)	1-10	10^2 - 10^3	5-50	10^{-4} - 10^{-3}	200 --
Chromium (Cr)	1-100	10^2 - 10^4	5-500	10^{-4} - 10^{-2}	100^e 1.5
Arsenic (As)	10-100	10^3 - 10^4	50-500	10^{-3} - 10^{-2}	500 3
Vanadium (V)	10-100	10^3 - 10^4	50-500	10^{-3} - 10^{-2}	50 2
Nickel (Ni)	10-100	10^3 - 10^4	50-500	10^{-3} - 10^{-2}	100 --
Lead (Pb)	10-100	10^3 - 10^4	50-500	10^{-3} - 10^{-2}	150 0.7
Manganese (Mn)	10-100	10^3 - 10^4	50-500	10^{-3} - 10^{-2}	5000 10
Zinc (Zn)	10^{-3} - 10^3	10^3 - 10^5	50-5000	10^{-3} - 10^{-2}	5000 --

^aThe gas containing all of the ash.^bBy ESP and wet FGD. 50% removal from mercury. 95% removal for others.^cOccupational safety regulation.^d24 hour average.^eFor Cr

TABLE 1-12. EXAMPLE DISSOLUTION TEST OF HAZARDOUS COMPONENTS FROM FLY ASH (mg/l) (Environment Agency, Japan)

Component	Type of Coal		Regulations	
	Domestic	Imported	Landfill	Ocean Dumping
Hg (organic)	ND ^a	ND ^a	ND ^a	ND ^a
Hg (total)	ND ^a	ND ^a	<0.005	<0.005
Cd	<0.018	ND ^a	<0.3	<0.1
Pb	<0.02	ND ^a	<3	<1
P (organic)	ND ^a	ND ^a	<1	<1
Cr ⁺⁶	<0.20	<0.10	<1.5	<0.5
As	<0.012	<0.38	<1.5	<0.5
CN	<0.01	ND ^a	<1	<1

^aNot detected

TABLE 1-13. SAMPLE FLY ASH COMPONENT DISSOLUTION TESTS (mg/l) (Environment Agency, Japan)

Component	Type of Coal		Emission Standards
	Domestic	Imported	
COD	0.7	1.6	120
BOD	1.6	—	120
Phenol	ND ^a	ND ^a	5
Cu	0.18	ND ^a	3
Zn	0.08	ND ^a	5
Mn	0.02	ND ^a	10
Cr (total)	ND ^a	0.18	2
F	0.11	1.02	15
PH	12.5	10.6	5.8 - 8.6

^aNot detected

efficiently caught by ESP and are relatively rich in heavy metal components. Baghouses may be able to catch the submicron particles more efficiently than ESPs. However, baghouses have not been used with utility boilers in Japan because of their high pressure drop and possible malfunctions.

SECTION 2

STATUS OF FLUE GAS DESULFURIZATION (FGD) IN JAPAN

2.1 FGD PLANTS AND BY-PRODUCTS

The treatment capacity of FGD plants in Japan increased rapidly from 1972 to 1976, reaching a total capacity of $100 \times 10^6 \text{ Nm}^3/\text{h}$ (32,000 MW equivalent) in 1976. Since then growth has been slower. In 1984, the total number of FGD plants was 1,400 with a total capacity of $130 \times 10^6 \text{ Nm}^3/\text{h}$ (42,000 MW equivalent). About 65% of the total capacity is associated with utility boilers, including 8,060 MW (27 plants) for coal-fired boilers. Most of the FGD plants constructed between 1977 and 1984 are for coal-fired boilers.

FGD plants built by major constructors are categorized in Table 2-1. The table identifies 481 FGD plants which represent a treatment capacity of about $116 \times 10^6 \text{ Nm}^3/\text{h}$. These plants account for 89% of the total FGD treatment capacity. About 119 plants use the wet lime/limestone process to produce a gypsum by-product; 72 plants use the indirect (dual-alkali type) or modified lime/limestone process and produce gypsum; 28 plants use the regenerable process and produce sulfuric acid, sulfur and ammonium sulfate; and 262 plants use sodium scrubbing to produce a sodium sulfite by-product for paper mills. In addition, there are over 900 small FGD plants with an average unit capacity of $15,000 \text{ Nm}^3/\text{h}$ (5 MW equivalent) using mainly sodium scrubbing to produce a sodium sulfite by-product for paper mills. The production capacity of FGD by-products is shown in Figure 2-1.

TABLE 2-1. FGD PLANTS BUILT BY MAJOR CONSTRUCTORS (Operational by the end of 1984)

Plant Constructor	Wet Lime/ limestone	Indirect/modified Lime/limestone	Numbers/capacity (1,000 Nm ³ /h)			Total
			H ₂ SO ₄ (M ₂) ₂ SO ₄	Na ₂ SO ₃ Na ₂ SO ₄	Na ₂ SO ₃ Na ₂ SO ₄	
Mitsubishi Heavy Industries (MHI)	44(29,174)	--	--	3(292)	--	47(29,446)
Hitachi Ltd./Babcock Hitachi	19(11,127)	--	2(590)	19(843)	--	40(12,560)
Ishikawajima-Harima H.I. (IHI)	20(8,633)	--	--	26(3,240)	--	46(11,873)
Kawasaki Heavy Industries (KHI)	2(174)	12(8,376)	--	7(256)	--	21(9,166)
Mitsubishi Kakoki (MKK)	5(506)	--	13(6,606)	3(530)	--	21(7,642)
Oji Engineering	--	--	--	32(6,239)	--	32(6,239)
Chiyoda Chemical Eng. Construct.	4(1,786)	13(4,375)	--	--	--	17(6,161)
Fujikasui Engineering	7(4,144)	--	--	18(1,716)	--	25(5,860)
Kurabo Engineering	--	5(558)	1(18)	102(3,912)	--	108(4,518)
Mitsui Miike Machinery (MMMC)	6(3,653)	--	1(520)	--	--	7(4,173)
Takishima Kikai (TSK)	1(80)	4(430)	1(110)	31(3,553)	--	37(4,173)
Ebara Corporation	1(26)	11(1,520)	--	10(2,006)	--	22(3,552)
Nippon Kokan (NKK)	3(197)	1(150)	2(1,990)	--	--	6(2,337)
Kobe Steel	--	6(1,925)	--	--	--	6(1,925)
Kureha Chemical	--	1(40)	--	7(1,380)	--	8(1,420)
Dowa Engineering	--	16(1,408)	--	--	--	16(1,408)
Shoden Engineering	--	3(1,002)	--	2(370)	--	5(1,372)
Mitsui Metal Engineering	4(1,006)	--	2(130)	--	--	6(1,136)
Sumitomo Chemical Engineering	--	--	5(748)	--	--	5(748)
Niigata Iron Works	2(265)	--	--	2(220)	--	4(485)
JGC Corporation	1(330)	--	1(125)	--	--	2(445)
Total	119(61,111)	72(19,784)	28(10,837)	262 (24,557)	--	481(116,289)

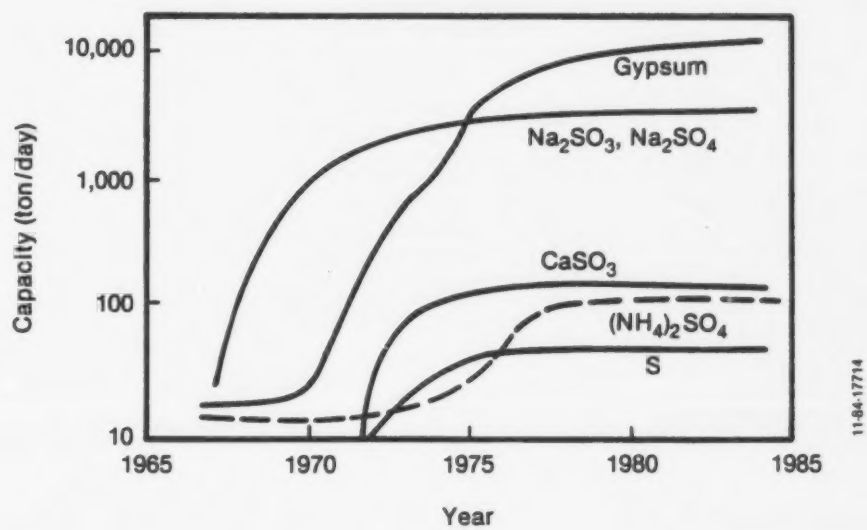


Figure 2-1. Production capacity of FGD by-products.

Gypsum is considered to be the most important by-product in Japan because the country has little natural gypsum. Technologies have been developed to produce nearly pure gypsum crystals that can be used in the production of wallboard, plaster, and cement. Gypsum was in oversupply in 1975 due to a rapid increase in the availability of FGD gypsum; this drove the selling price to almost zero (Figure 2-2). However, since 1977, there has been a shortage of gypsum because of an increase in demand and a concomitant decrease in the production of gypsum from wet process phosphoric acid production. By-product gypsum currently can be sold for 5,000 yen/ton. All FGD plants for utility coal-fired boilers produce salable gypsum as a by-product. Only a few FGD plants produce calcium sulfite sludge for disposal.

Sodium scrubbing which produces a sodium sulfite by-product is the simplest FGD system. Many sodium scrubbing plants were constructed between 1968 and 1976 because the sodium sulfite by-product could be sold to paper mills. An oversupply of sodium sulfite resulted, which led numerous small FGD plants to switch to the production of a waste liquor containing sodium sulfite and sulfate. Recently, several magnesium scrubbing plants have been constructed because magnesium hydroxide, the absorbent used to by-produce the waste magnesium sulfate liquor, is cheaper than the sodium hydroxide or carbonate used in other processes.

Most regenerable process FGD plants were constructed between 1972 and 1976, when wet limestone-gypsum FGD process technology was not yet well established, and an oversupply of gypsum was anticipated. Those processes have not been used widely because of the high cost of the technology and the oversupply of by-products.

2.2 WET LIME/LIMESTONE GYPSUM PROCESS

Most new large FGD plants in Japan use a wet limestone process which yields gypsum as a by-product. This process is preferred for the following reasons: 1) limestone is abundant in Japan; 2) the gypsum by-product is salable; 3) the process has become quite reliable, and the plants have been

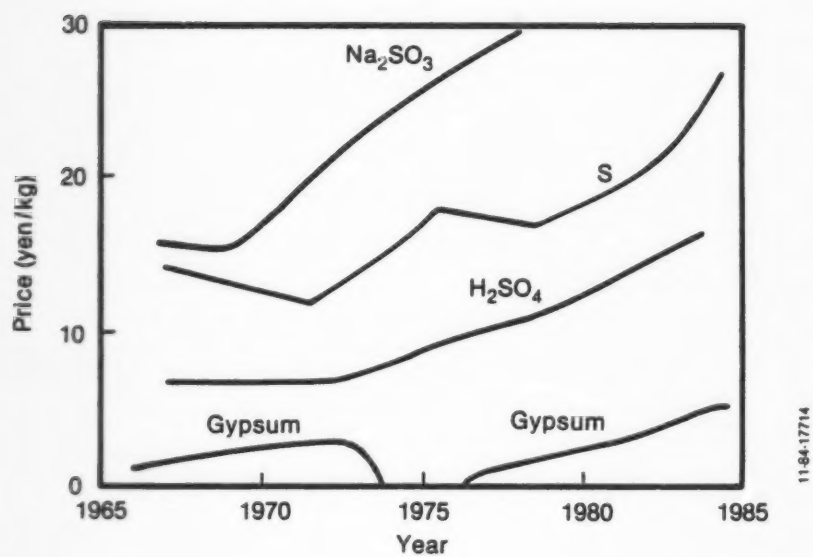


Figure 2-2. Price of FGD byproducts.

operated with over 99% reliability; 4) the process has a relatively low cost compared with other FGD technologies; 5) the small amount of coal fly ash contained in the gypsum by-product does not significantly affect the gypsum quality; 6) over 95% of SO_2 can be removed by using finely ground limestone (325 mesh, 90% pass); and 7) limestone is much less expensive than lime. Although the scrubber size and the L/G ratio may be larger with limestone than with lime, the overall process cost is lower with limestone.

A flowsheet and a material balance for a typical limestone-gypsum process for a 500 MW coal-fired boiler is shown in Figure 2-3. Flue gas is cooled by a gas-gas heater and introduced into a prescrubber. There it is washed with a water spray for further cooling and dust removal. The gas is then introduced into a scrubber through a mist eliminator. In the scrubber, 90-98% of SO_2 is absorbed by a slurry containing limestone, calcium sulfite, and gypsum. The treated flue gas is sent to the gas-gas heater through a mist eliminator. Slurry leaving the scrubber at pH 5-6 contains calcium sulfite and gypsum with a small amount of limestone. Calcium sulfite is oxidized to gypsum through the use of air bubbles in an oxidizing tower. Usually a small amount of sulfuric acid is added to the sulfite slurry to promote the oxidation process. Gypsum is centrifuged to yield a product containing less than 10% moisture. Wastewater is discharged from both the prescrubber and the scrubber system and is treated in a wastewater treatment system for neutralization, removal of heavy metals, suspended solids, COD, etc. Variations of the limestone-gypsum process also are in use; they are described in Section 3 of this report.

2.2.1 Recent Improvements

Since technology has already been developed to prevent scaling, a major problem in the wet lime/limestone FGD process, recent research efforts have focused on making the process more cost-effective through reduced power consumption and investment costs (1, 3). Table 2-2 summarizes investment costs and power consumption for limestone FGD processes. The power consumption of FGD plants constructed before 1978 for coal-fired boilers reaches 2.4-2.6% of the power generated by the boiler. Most of this power is

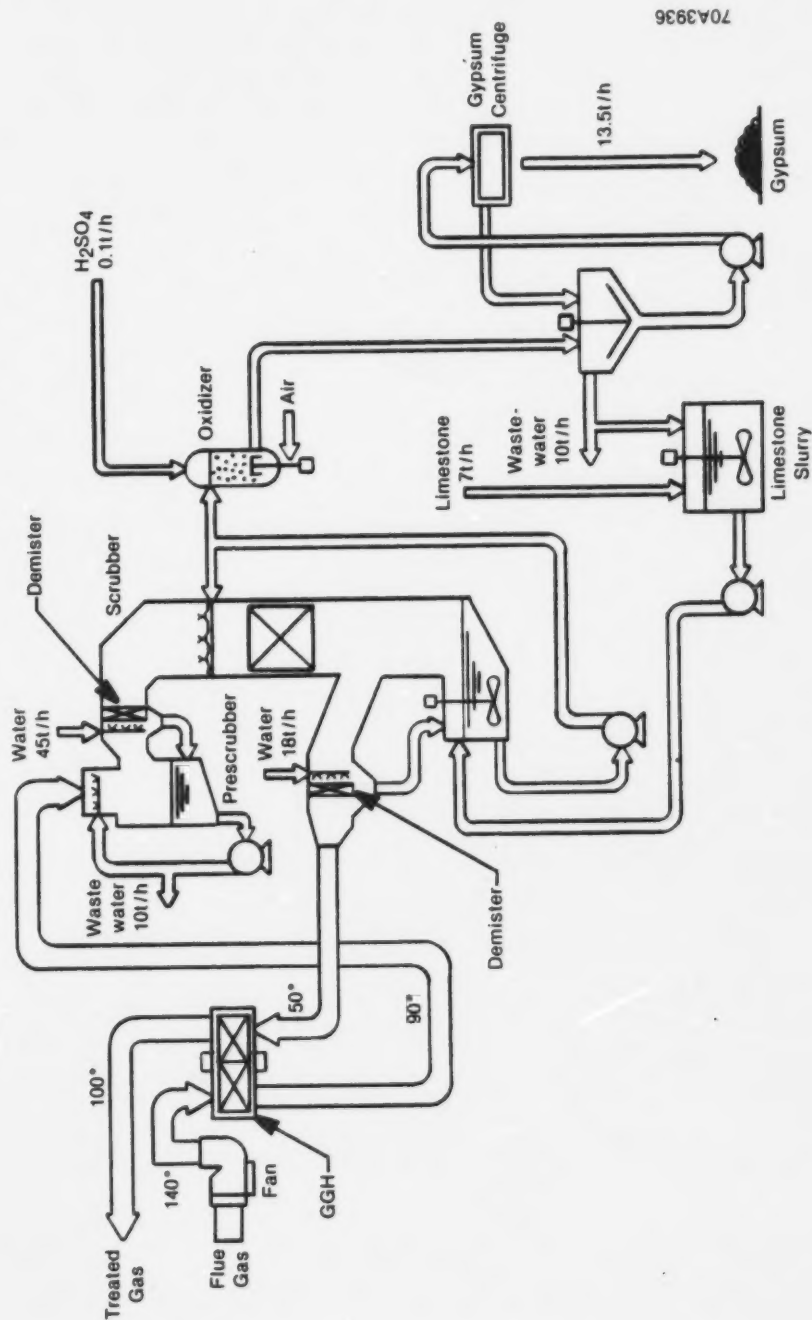


Figure 2-3. Flowsheet and material balance of limestone-gypsum FGD process. (500 MW coal-fired boiler. Inlet SO₂ 1,000 ppm)

TABLE 2-2. INVESTMENT COST AND POWER CONSUMPTION OF FGD PLANTS FOR COAL-FIRED BOILERS (Limestone-gypsum process)

Power Company	Station Name	Capacity (MW)	New or retro-fit	Year completed	Maker	Scrubber Type	Investment Cost (yen/KW)	Power consumption (%)
EPDC	Takasago	250	R	1975	MMMC ^a	Venturi	18,600	2.6
EPDC	Isogo	265	R	1976	IHI ^b	Venturi	20,750	2.4
EPDC	Takehara	250	R	1977	BHK ^c	P. P. ^d	28,800	2.6
Chugoku	Shimonoseki	175	R	1979	MHI ^e	Packed	34,300	2.3
Hokkaido	Tomato	350 ± 1/2	N	1980	BHK	P. P. ^d	-	1.7
Tohoku	Sendai	175	R	1983	BHK	Spray	40,000	-
EPDC	Takehara	700	N	1983	IHI	Spray	20,000	2.1
Joban	Nakoso	600 ^g	N	1983	MHI	Packed	21,000	1.8
Kyushu	Minato	175	R	1983	MMMC	Spray	24,000	2.0
Toyama	Toyama	200	R	1984	Chiyoda	JBR ^f	19,000	1.6

^aMitsui Miike Machinery Co.

^bIshikawajima-Harima Heavy Industries.

^cBabcock-Hitachi K. K.

^dPerforated plate.

^eMitsubishi Heavy Industries.

^fJet bubbling reactor.

^gWith coal, 30; oil, 70.

consumed by the fan required because of the pressure drop caused by the FGD system. Figure 2-4 shows that the power consumption of such a fan in an FGD plant using a perforated plate scrubber was reduced considerably simply by enlarging the openings in the perforated plate. Although pump power consumption increased slightly when the L/G ratio was increased to maintain SO₂ removal efficiency at the same level, total power consumption was lowered (EPDC's Takehara plant constructed in 1977 vs Hokkaido's Tomato plant).

Recently, the use of a spray tower absorber has become popular because of the slight pressure drop and the absence of packings which might cause scaling. Through the use of efficient multi-stage spray nozzles, over 95% of the SO₂ can be removed with a limestone slurry (see Figure 2-5). MHI continues to use the low-pressure drop-type, plastic-grid-packed tower

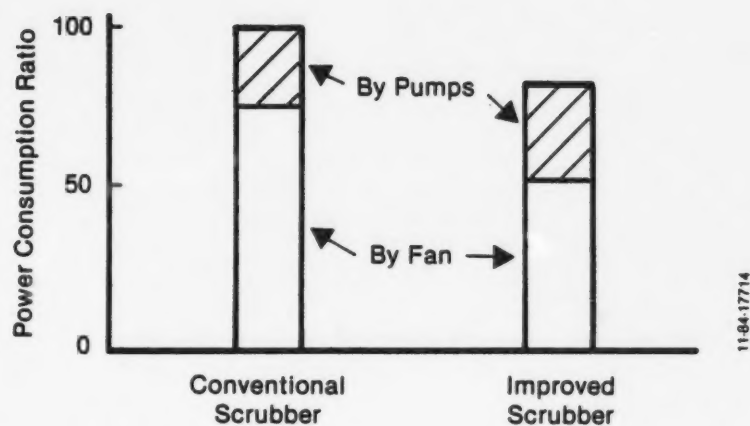


Figure 2-4. The effect of scrubber improvements on power consumption.

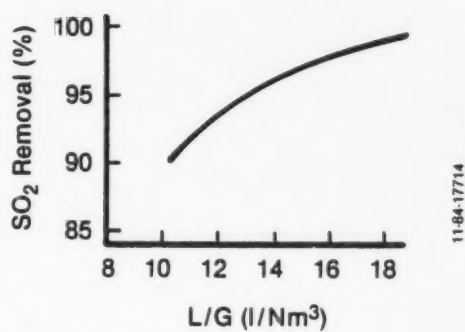


Figure 2-5. L/G vs. SO₂ removal efficiency (4).
(spray tower, pH=5.6; inlet SO₂=800 ppm)

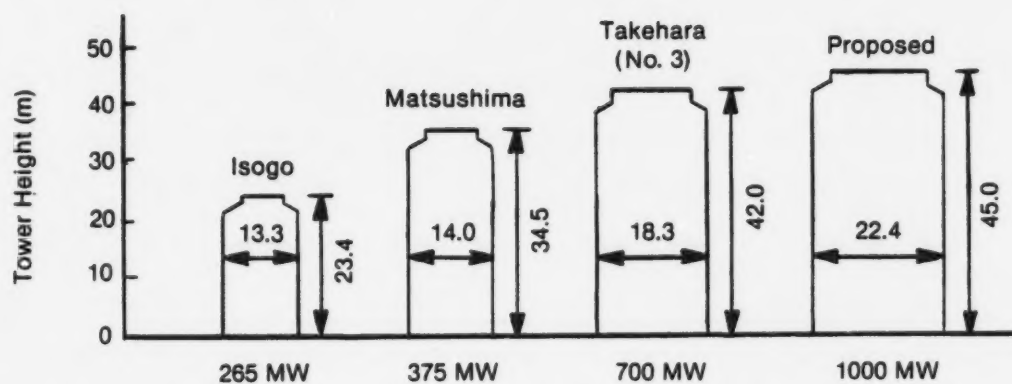
absorber. Chiyoda has recently developed an entirely new scrubber that consumes less power without slurry recirculation. Computer control which reduces the power consumption of the FGD operation also has become popular. As a result of these innovations, the power consumption of FGD plants completed since 1980 has been lowered to 1.6-2.1%.

The investment cost for lime/limestone FGD systems in yen/KW increased from 18,600 in 1975 to 34,300 in 1979 mainly due to inflation. However, most of the FGD plants constructed in 1983 and 1984 cost between 19,000 and 24,000 yen/kW. These reduced investment costs may be due to: 1) technology improvements which allow a plant to be simplified without reducing its reliability and SO₂ removal efficiency, 2) construction of many FGD plants which allows plant constructors to realize their return on R&D expenses, and 3) demand for less costly plants particularly because of the recent economic downturn. Overall, reduced investment costs and power consumption have made FGD considerably less costly.

A comparison of spray tower absorbers with different capacities is shown in Figure 2-6. The absorber at the Takehara No. 3 plant, which may be the largest in the world, treats all of the gas from a 700 MW boiler in a single absorber. Normally, FGD plants in the U.S. have several scrubbers in parallel, including a spare scrubber. Japanese FGD plants, on the other hand, do not require a spare scrubber because the main scrubber can be operated continuously for over a year. Once a year, utility boilers shut down for maintenance for about a month. Problems such as scaling or corrosion can be taken care of during this shutdown period. As a precaution, power stations usually store low-sulfur oil, so that if an FGD plant has a serious problem, it can be shut down while the boiler is operated by burning low-sulfur oil. However, this situation rarely occurs; FGD plants in Japan have been operated with over 99% reliability.

2.2.2 Computer Control

Limestone feed in FGD systems usually is controlled by the inlet SO₂ mass loading. New large FGD plants have computer control systems to improve their



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Figure 2-6. Comparison of spray tower scrubbers with varying capacities (4). (Ishikawajima-Harima Heavy Industries)

operation. For example, Mitsubishi Heavy Industries (MHI) has developed computer software called ASC-1000 (Advanced Scrubber Control) for limestone scrubber systems. The system has the capability to perform on-line forecasting of operating conditions, thus enabling the plant operator to reduce limestone and power consumption by responding quickly to changes in inlet conditions. The operation of the software and its response to changes in the boiler load and inlet SO_2 concentrations are shown in Figure 2-7 and 2-8. This software system has been used at the FGD plant for the No. 8 boiler (600 MW) at the Nakoso Power Station.

2.2.3 Estimated FGD Cost

The estimated FGD cost for a 500 MW coal-fired boiler equipped with a gas-gas heater is shown in Table 2-3. The annualized FGD cost is about 1.3 yen/kWh. About 70% of the cost consists of the fixed cost with a 7-year depreciation schedule. When the annualized cost is based on a 15-20 year depreciation schedule, as is commonly done in the U.S., the result is a considerably lower yen figure.

2.3 INDIRECT AND MODIFIED FGD LIME/LIMESTONE PROCESSES

Indirect (dual-alkali type) and modified lime/limestone FGD processes are shown in Table 2-4. These processes have been developed to compensate for some of the weaknesses of wet lime/limestone processes. More specifically, the indirect and modified processes are designed to incorporate these features: 1) no scaling; 2) high SO_2 removal efficiency; 3) lower power consumption than the limestone process; 4) easy plant operation; and 5) no need for sulfuric acid. On the other hand, most indirect processes are complex and require a high investment cost. In addition, these processes have become less attractive as the wet lime/limestone process has been improved, especially with regard to decreased scaling and increased SO_2 removal efficiency.

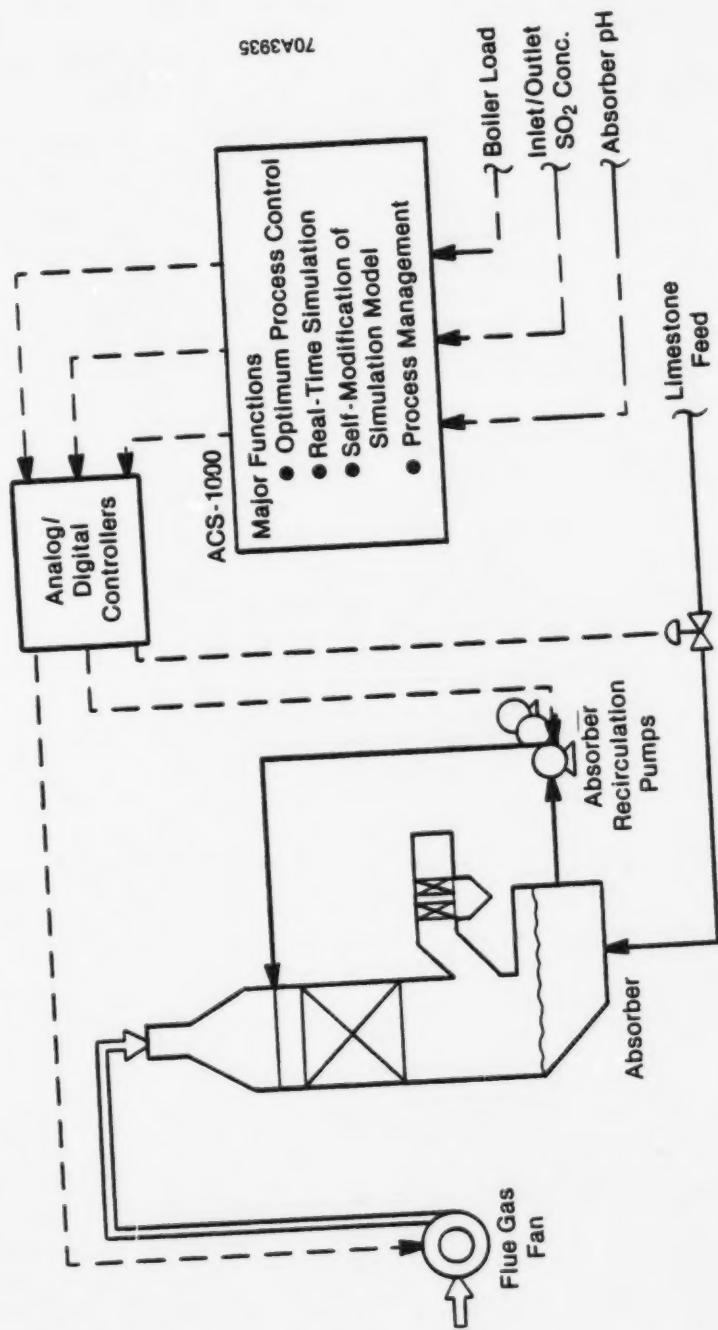


Figure 2-7. ACS-1,000 control system (MHI).

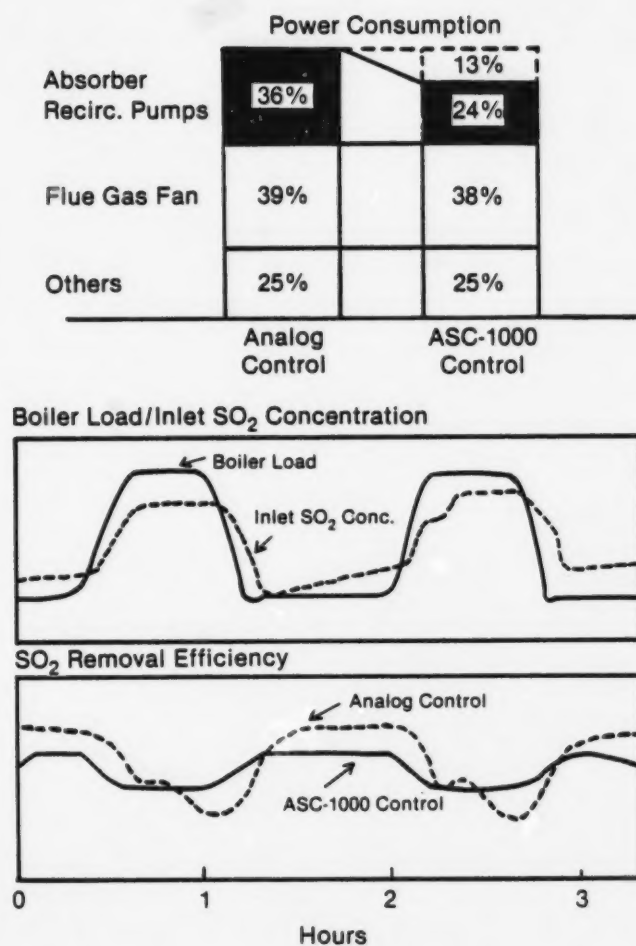


Figure 2-8. Example of MHI's ASC-1,000 control of an FGD system and resultant power savings.

TABLE 2-3. ESTIMATED FGD COST IN 1984 (Assuming a new 500 MW coal-fired boiler, limestone-gypsum process, 70% boiler utilization, 3,066,000 MWh/year, inlet SO₂ 1,000 ppm, 90% SO₂ removal, 7 years depreciation, 10% interest.)

Investment Cost (including gas-gas heater)	10 billion yen (20,000 yen/kW)
Annual Cost (millions of yen)	
Fixed Cost (25% of investment)	2,500
Power (1.8% of power generated, 15 yen/kWh)	840
Labor (8 persons)	50
Limestone (43,000 tons) and chemicals	350
Others	<u>800</u>
Total	4,540
Gypsum (84,000 tons, 3,000 yen/ton)	<u>-250</u>
Total	4,290
Annualized Cost	1.40 yen/kWh

TABLE 2-4. INDIRECT AND MODIFIED LIME/LIMESTONE PROCESSES
(Gypsum by-product)

Developer	Absorbent	pH	Precipitant
<u>Indirect Process</u>			
Kureha-Kawasaki	Na_2SO_3	7	CaCO_3
Showa Denko (Shoden Eng.)	Na_2SO_3	7	CaCO_3
Tsukishima Kikai	Na_2SO_3	7	CaO
Kurabo	$(\text{NH}_4)_2\text{SO}_3$	7	CaO
Nippon Kokan	$(\text{NH}_4)_2\text{SO}_3$	7	CaO
Dowa	$\text{Al}_2(\text{SO}_4)_3$	3.5	CaCO_3
Chiyoda (CT 101)	Dilute H_2SO_4	1	CaCO_3
<u>Modified Process</u>			
Kawasaki H. I.	$\text{CaO} + \text{MgO}$	5.5	-
Kureha	$\text{CaCO}_3 + \text{Sulfo-succinate}$	5	-
Kobe Steel	$\text{CaO} + \text{CaCl}_2$	5	-
Nippon Steel	Convertor slag	5	-

No new indirect process FGD plants have been constructed since 1980 except for ones utilizing the Dowa process. Chiyoda has discontinued its dilute sulfuric acid absorption process because of its high cost and instead has developed a unique wet limestone-gypsum process which is less costly. The latter process will be described in Section 3.5.

Among the modified lime/limestone processes, the Kawasaki lime-magnesium process has undergone further development and three new lime-magnesium plants for coal-fired boilers have been constructed in West Germany and Japan. Two plants using the Dowa aluminum sulfate process were completed in 1983. In 1982, one Kureha organic acid process plant and one Nippon Steel convertor slag process FGD plant began operation. All of these new plants are characterized by low investment costs and reduced power consumption. They are described in more detail in Section 3 of this report.

2.4 REGENERABLE AND DRY FGD PROCESSES

2.4.1 Regenerable FGD Processes

The major regenerable FGD process used in Japan is the Wellman-Lord. As shown in Table 2-1, Mitsubishi Kakoki (MKK) has constructed 15 Wellman-Lord plants and Sumitomo Chemical Engineering (SCEC) has built an additional five. Sodium scrubbing is followed by heating of the resultant sodium bisulfite solution. From this solution, concentrated SO_2 gas is recovered for use in production of sulfuric acid at 15 plants. At three plants installed at oil refineries, the recovered SO_2 is introduced into existing Claus furnaces where it reacts with H_2S to produce elemental sulfur. At one plant, the recovered SO_2 is used in a chemical process.

The Wellman-Lord requires a high investment cost and also has become costly because of the large amount of energy required for evaporation of the process liquor. Recently, improvements have been made in the evaporator and in the recovery of sodium sulfate formed by the oxidation of sodium sulfite. However, no new Wellman-Lord plants have been constructed since 1978.

Mitsui Miike Machinery Co. constructed a magnesium scrubbing plant at Idemitsu Nosan's Chiba refinery (3). At the plant, the magnesium sulfite-sulfate that is produced is filtered, dried and calcined to recover concentrated SO_2 gas. The gas is then introduced into an existing Claus furnace to produce elemental sulfur. Drying and calcination problems have been solved so that the plant can be operated with high reliability. However, high energy consumption due to drying and calcination has made the process costly. In the past, there were two other magnesium scrubbing plants recovering concentrated SO_2 for sulfuric acid production, but they were shut down several years ago.

A number of ammonia scrubbing plants, including two large ones belonging to Nippon Kokan and several small ones, produce ammonium sulfate as a by-product. At the Nippon Kokan plant, SO_2 from oil-fired boilers and NH_3 in coke oven gas are combined to produce sulfate. Plant operation has been successful. An oversupply of ammonium sulfate, however, has prevented the construction of new ammonia scrubbing plants.

2.4.2 Dry FGD Processes

Dry processes attracted attention in Japan in the early 1970s and again in the early 1980s mainly because they don't require any gas reheating, and therefore produce no wastewater. The only dry process in commercial use is the activated carbon (activated coke) process. At Tokyo Electric's Kashima Station, a portion of the gas (150 MW equivalent) from an oil-fired boiler is treated with activated carbon in a fixed bed to remove about 80% of the SO_2 . The SO_2 -loaded carbon is washed with water to produce a dilute sulfuric acid which is treated with limestone to produce salable gypsum. This process produces a considerable amount of wastewater and has not become popular. At another carbon adsorption plant, the dilute sulfuric acid obtained by washing the carbon is sprayed into the inlet flue gas at 170°C . This concentrates the acid to 60%. The by-product acid is used at a chemical plant.

The activated coke FGD process has been tested for commercial use. In this process, SO_2 -loaded carbon is heated to produce a concentrated SO_2 gas for use in elemental sulfur production. The process has not been commercialized yet, however, primarily due to its high carbon consumption.

A dry process was used commercially in the past is the Shell copper oxide process. A copper oxide plant was operated for several years and then was shut down, possibly because of the high operation cost.

A semi-dry process similar to the spray dryer process used in the U.S. was studied in Japan in the early 1970s. A sodium scrubbing plant has been operated using a spray of sodium hydroxide liquor to by-produce powdery sodium sulfite which can be sold to paper mills. The low quality product contains sodium sulfate and carbonate. This process has not become popular.

The FGD spray dryer system with lime scrubbing will not be applied in Japan because of the by-product disposal problem. Spray dryer systems also may not be able to achieve the 95% removal efficiency required of new FGD plants in Japan.

Two dry simultaneous SO_x and NO_x removal processes currently are being developed for commercial application. These processes are described in Section 6.

2.5 CONSTRUCTION MATERIALS

Most of the FGD plants constructed recently use carbon steel lined with glass-filled resin for scrubber vessels, tanks, ducts, etc. A rubber lining is sometimes used for pumps, hydrocyclone and other system components. The linings generally perform well. Well fabricated linings prevent corrosion and are crucial to highly reliable FGD operation.

Stainless steel use is limited to parts such as pumps and agitators. Spray nozzles usually are made of silicon carbide. Stacks are made of stainless steel or carbon steel lined with bricks or gunite.

2.6 GAS REHEATING AND WASTEWATER TREATMENT

2.6.1 Gas Reheating

Until 1978, flue gas leaving a wet FGD plant was reheated exclusively by low-sulfur oil firing. Reheating the gas to 110-130°C consumed 3-4% of the energy needed to run the boiler and therefore was quite costly. FGD plants for coal-fired boilers completed since 1979 all use a rotating Ljungstrom gas-gas heater. It has been found that coal ash which is deposited in the gas-gas heater (GGH) is easily removed by soot blowing, while the ash from oil-firing is difficult to remove. Although the GGH is quite costly--about 10% of the FGD investment cost--it is a more economical method of gas reheating than low-sulfur oil-firing. GGH requires no oil-firing and the FGD inlet gas is cooled, resulting in decreased water consumption.

One of the problems associated with the GGH is gas leakage. As shown in Figure 2-9, the fan usually is located at the A position, causing 2-5% of the untreated flue gas to leak into the cleaned gas. This leakage reduces both the SO₂ and dust removal efficiencies. Placing the fan at the B or C position causes the cleaned gas to leak into the untreated gas, thus preventing any decrease in SO₂ or dust removal efficiencies.

Placing the fan in the B or C positions may be preferable when a very high SO₂ removal efficiency (over 97%, for example) is required. The EPDC's Takehara No. 3 plant constructed by IHI uses the B position, while Mitsui Aluminum's No. 2 plant at Omuta and Mitsui Coal's Miike plant use the C position. However, locating the fan at the B or C position is more costly than placing it at the A position. Corrosion-resistant material must be used in the B and C positions due to the acid condensation that occurs at the low temperature in that part of the heater. MMC prefers the C position because the actual gas volume is smaller at the low temperature and the fan can be water-washed during the operation.

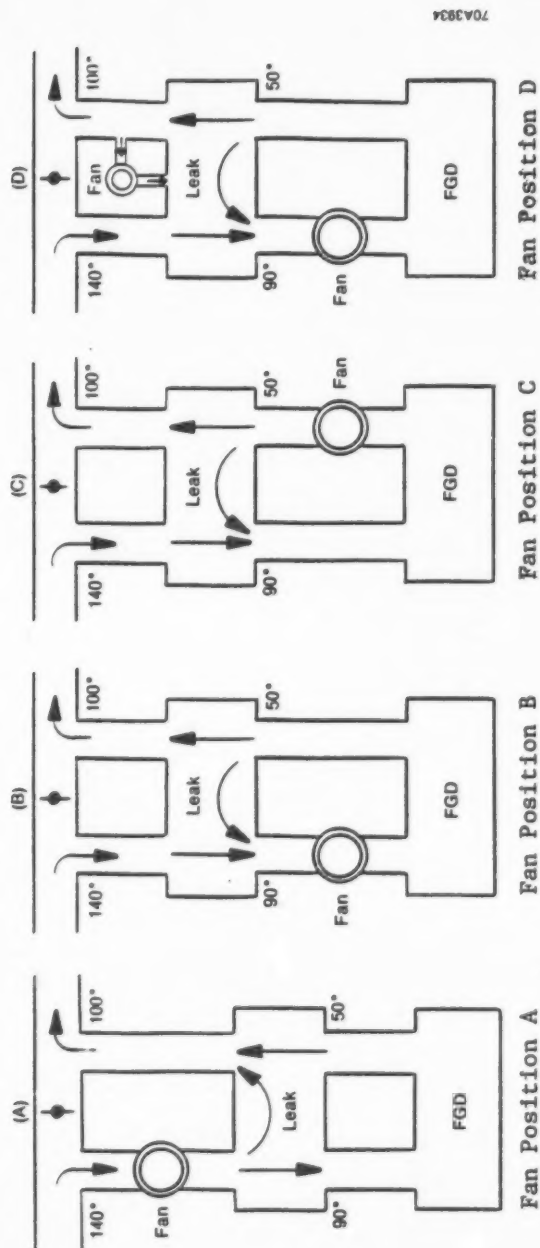


Figure 2-9. Gas leakage in a rotary gas-gas heater.

Various improvements have been made in gas-gas heaters in order to reduce gas leakage. For example, the GGH at Joban Power's Nakoso Station has a fan at the A position with less than 2% leakage. Even with the fan in the B and C positions, a slight decrease in SO_2 and dust removal efficiencies cannot be prevented because the gas containing SO_2 and dust remains between the heating elements and is blown into the treated gas. To prevent this, a scavenging fan has been installed at the Takehara station, as shown in D of Figure 2-9.

Another problem with the GGH is dust deposition. Even with soot blowing, a small amount of dust deposits in the GGH, increasing the pressure drop. The amount of dust deposited is directly related to the SO_3 and dust content of the flue gas. The higher the SO_3 content, the greater the amount of deposits. With a coal-fired flue gas which usually contains 3-5 ppm SO_3 , the pressure drop increases only slightly when the dust content is less than 50 mg/Nm^3 . The pressure drop increases considerably when the dust content is about 200 mg/Nm^3 , necessitating a water wash two or three times a year. The washing may take two to four days including cooling and drying time. When the dust content is even higher, the deposits are softer and may be removed relatively easily by soot blowing, thus requiring less water washing.

Non-leak gas-gas heaters also have been tested. At Tokyo Electric's Yokosuka Station, two FGD plants for 265 MW COM-fired boilers are near completion. One of the plants uses a multi-tube gas-gas heater developed by MHI, while the other uses a BHK heat pipe heater.

2.6.2 Wastewater Treatment

The composition of wastewater from coal-fired boilers is shown in Table 2-5. Figure 2-10 illustrates a typical wastewater treatment system for a FGD plant.

Coagulation precipitation by lime addition is used to remove fluoride, heavy metals, and suspended solids. Most large FGD plants in Japan also are required to remove COD that is mainly in the form of dithionate, ($\text{S}_2\text{O}_6^{-2}$). Dithionate removal is complicated, requiring ion-exchange resin adsorption, pH adjustment, and disposal of the adsorbed COD. Dithionate is formed from

TABLE 2-5. pH AND COMPOSITION (Mg/liter) OF WASTEWATER FROM FGD UNITS ON COAL-FIRED BOILERS PRIOR TO TREATMENT (Environment Agency)

pH		1 ~ 5
Chemical oxygen demand	(COD)	30 ~ 100
Suspended solids	(SS)	2,000 ~ 3,000
Fluoride	(F)	500 ~ 600
Iron	(Fe)	100 ~ 500
Manganese	(Mn)	10 ~ 30
Magnesium	(Mg)	200 ~ 1,000
Aluminum	(Al)	100 ~ 1,400
Chloride	(Cl)	3,000 ~ 8,000
Total nitrogen ^a	(N)	100 ~ 600

^aWhen SCR of NO_x is not applied.

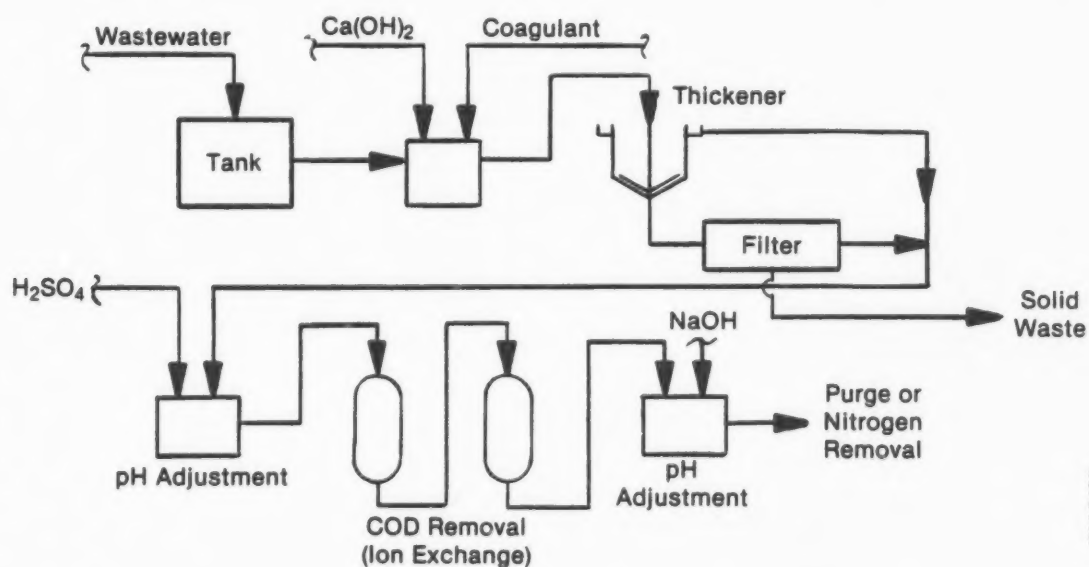


Figure 2-10. Typical FGD wastewater treatment system.

sulfite and is generally more concentrated with sulfite-rich scrubber liquor. A relatively large amount of dithionate is formed by the Wellman-Lord process in which the sulfite liquor is heated. With the Chiyoda JBR process, the sulfite concentration is limited, due to the air that is bubbled into the reactor. For this reason, only a small amount of dithionate is formed.

FGD wastewater also contains a small amount of nitrogen compounds derived mainly from the NO_x in the flue gas. A small portion of NO_x , possibly 1-3%, is absorbed in the scrubber liquor to form nitrate and N-S compounds such as imidodisulfamate. Generally there is not enough nitrogen in the wastewater to warrant its removal. When SCR is applied, the treated gas contains a small amount of unreacted NH_3 which is caught by the scrubber. Normally the ammonia concentration in the wastewater is low (below 100 mg/Nm^3) due to the low (less than 3 ppm) concentration of unreacted NH_3 , so ammonia removal is unnecessary. EPDC's Takehara plant has a biological ammonia removal facility for safety reasons.

2.7 PARTICULATE REMOVAL

Electrostatic precipitators (ESPs) are widely used to remove particulates from a large amount of gas. For relatively small amounts of gas, baghouses are used instead. Baghouses are not yet used for large amounts of gas, such as that produced by coal-fired utility boilers, because of their high pressure drop and the possibility of malfunction. Cold-side ESPs generally are used with coal-fired boilers except for 3 hot-side ESPs constructed between 1979 and 1982. Although ESPs for coal-fired boilers remove 96-99.8% of particulates, an FGD system also is used for the additional particulate removal required (Figure 2-11). Between 0.1 and 1 g/Nm^3 of the dust at the ESP outlet is reduced to 0.02 - 0.1 g/Nm^3 by the FGD system. In order to obtain a higher dust removal efficiency from a scrubber system, a larger L/G ratio must be used, which, in turn, causes a higher pressure drop (Figure 2-12). To increase the dust removal efficiency of an ESP, a larger ESP and higher

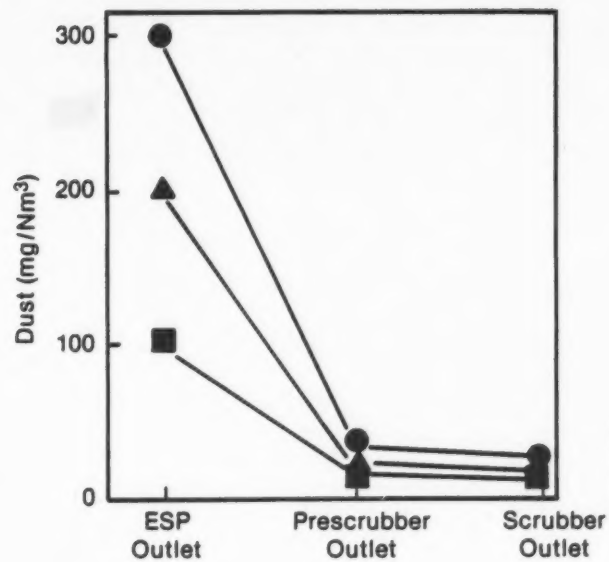


Figure 2-11. Dust removal by ESP and FGD.

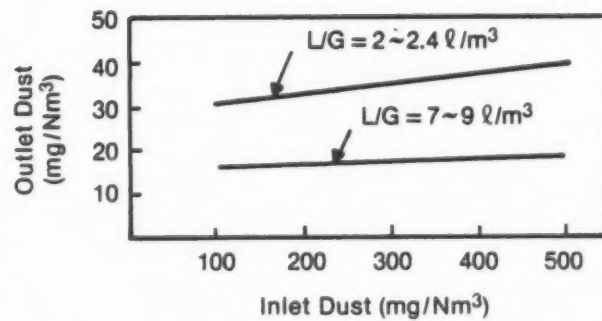


Figure 2-12. Dust removal by FGD with different L/G Ratios (4).

power consumption are required, although the removal efficiency also depends on the composition of the fly ash.

Figure 2-13 shows dust removal costs for reducing particulates to 0.05 g/Nm^3 using the combination of an ESP and a scrubber system. With a decrease in ESP efficiency, the investment depreciation costs decrease, but the scrubber system must be operated with a high dust removal efficiency, thereby consuming a large amount of power. The total cost may be the lowest when the ESP outlet particulate concentration is about 0.2 g/Nm^3 .

A small portion of the dust leaving the ESP is deposited in the gas-gas heater (GGH). When the GGH inlet dust concentration is $0.1\text{--}0.2 \text{ g/Nm}^3$, dust deposition in the GGH is considerable, even with soot blowing, and the GGH may have to be water-washed two or three times a year. Therefore, new plants for coal-fired boilers use an efficient ESP to reduce particulates below 0.1 g/Nm^3 so that the GGH can be operated continuously for a year.

Figure 2-14 shows the particle size distribution of fly ash from pulverized coal combustion at three stages of dust removal. The dust concentration at the GGH inlet is 227 mg/Nm^3 , with particles ranging in size from 0.2 to $3 \mu\text{m}$. These sizes indicate that particulates larger than $3 \mu\text{m}$ are caught by the ESP. In the scrubber and in the mist eliminator within the scrubber, the dust is reduced to 61.7 mg/Nm^3 and most of the particles between 1.5 and $3 \mu\text{m}$ are removed. With an efficient mist eliminator located between the scrubber and the GGH, particulates are further reduced to 11.9 mg/Nm^3 , with most of the remaining particles ranging from 0.2 to $1 \mu\text{m}$.

McElroy found that particles around $0.2 \mu\text{m}$ in size are relatively rich in heavy metals such as arsenic and zinc (6). Those elements and their compounds are vaporized at high combustion temperatures and condense on cooling to form very fine particles. The behavior of these fine particles merits further study, although they may not have an adverse effect on human health because they exist in such small quantities (Table 1-11).

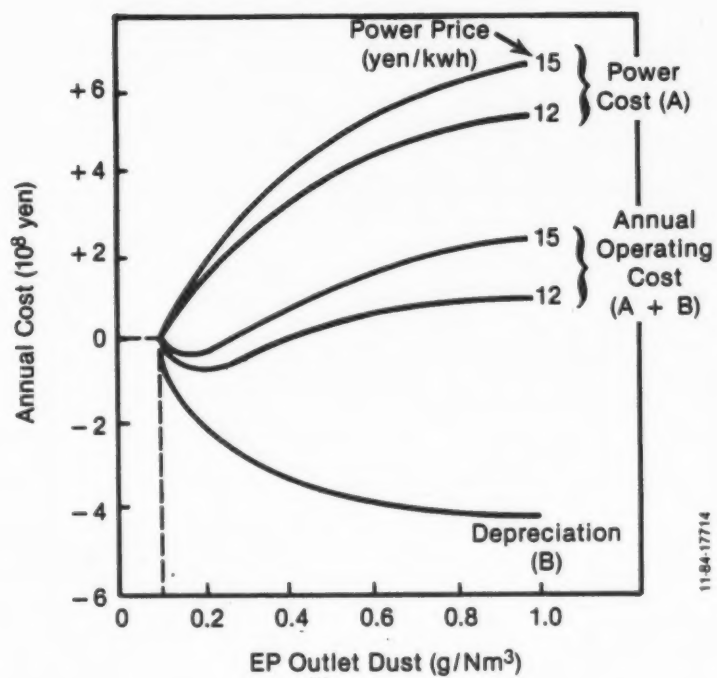
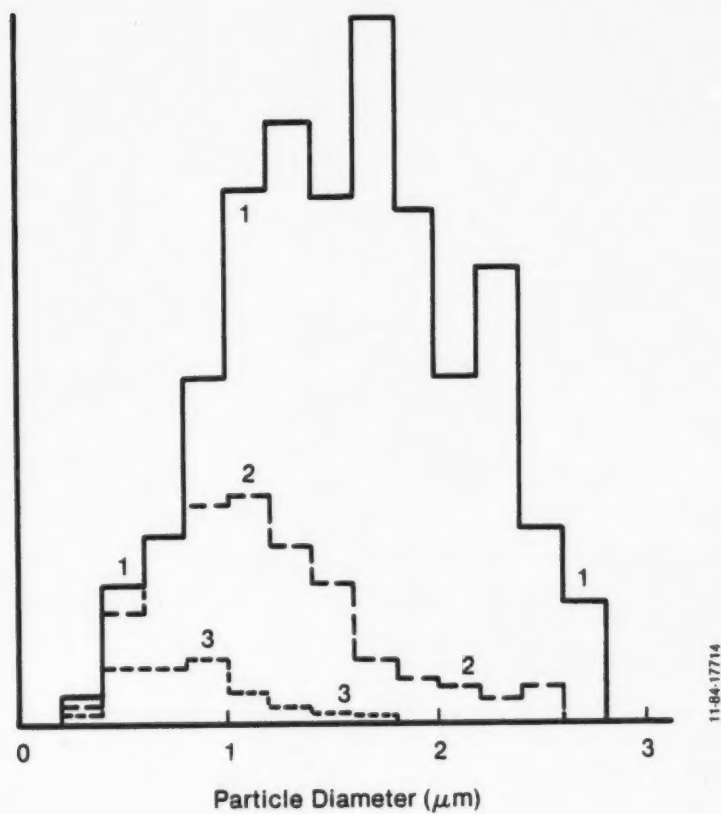


Figure 2-13. Cost of dust removal by ESP and FGD (1,000 MW coal-fired boiler. Outlet dust 50 mg/Nm^3) (5).



- 1 Gas-Gas Heater Inlet (Dust 227mg/Nm³)
- 2 Scrubber Outlet (Dust 61.5mg/Nm³)
- 3 Mist Eliminator Outlet (Dust 11.9mg/Nm³)

Figure 2-14. Particle size distribution of dust at three stages of removal (4).

2.8 FGD FOR COAL-FIRED BOILERS

Most coal-fired boilers in Japan have an FGD plant to remove 85-95% of SO_2 . Table 2-6 lists major coal-fired boilers and their FGD and SCR plants. In addition, there may be more than 10 smaller coal-fired boilers using low-sulfur coal without FGD. All of the coal-fired boilers have ESPs to reduce particulates to $50-800 \text{ mg/Nm}^3$ -- a removal efficiency of 96-99.8%. All of the FGD plants use the wet lime/limestone process, except Shikoku Electric's Saijo plant which uses a modified lime process.

FGD processes for coal-fired boilers can be divided into six types as shown in Tables 2-6 and 2-7 and Figure 2-15. Type I produces a disposable sludge consisting mainly of calcium sulfite. This type of FGD system is simple and is popular in the U.S. where there is adequate land for sludge disposal. Only one Type I plant has been operated in Japan because of the lack of space for sludge disposal.

Other types of FGD processes produce by-product gypsum by oxidation of calcium sulfite with air. To obtain salable gypsum of high purity, complete oxidation must be ensured by lowering the pH of the calcium sulfite slurry leaving the scrubber from 5.5-6 to 4-4.5 prior to the oxidation step. Type II FGD systems use an extra scrubber to lower the pH by further SO_2 absorption and add sodium hydroxide or sulfate to promote SO_2 absorption and oxidation. Type II systems are in operation at three FGD plants. Type I and II plants have been constructed by Mitsui Miike Machinery Co. (MMMC).

In Type III FGD plants the pH of the slurry is lowered by addition of a small amount of sulfuric acid. Type III systems require lower investment costs than Type II plants, and may be preferable where sulfuric acid is available for a reasonable price. The Type III system often uses a sodium compound to promote the reactions. Ishikawajima Harima Heavy Industries (IHI) and Babcock Hitachi K.K. (BHK) have constructed four Type III system plants,

TABLE 2-6. FGD AND SCR UNITS FOR COAL-FIRED BOILERS
(Larger than 75 MW) TO BE COMPLETE BY 1987

Owner	Plant Site	Boiler (MW)	FGD			SCR		
			Const- ructor	Comp- leted	Type	Const- ructor	Comp- leted	Type
Mitsui Aluminum	Omuta	156	MMMC ^a	1972	I	-	-	-
Mitsui Aluminum	Omuta	175	MMMC	1975	II	-	-	-
EPDC	Takasago	250 x 2	MMMC	1975, 76	II	-	-	-
EPDC	Isogo	265 x 2	IHI ^b	1976	III	-	-	-
EPDC	Takehara	250	BHK ^c	1977	III	BHK, KHI ^d	1980	L ^e
EPDC	Takehara	700	IHI	1983	III	BHK	1983	L
EPDC	Matsushima	500 x 2 ^f	IHI, BHK	1981	IV	-	-	-
EPDC	Ishikawa	156 x 2	IHI	1986	III	-	-	-
Chugoku Elec.	Shimonoseki	175	MHI ^g	1979	IV	MHI	1980	H ^h
Chugoku Elec.	Shin-Ube	75 x 2	MHI	1982	IV	MHI	1982	H
Chugoku Elec.	Shin-Ube	156	MHI	1982	IV	MHI	1982	H
Chugoku Elec.	Misushima	125 & 156	BHK	1984	IV	BHK	1984	H
Chugoku Elec.	Shin-Onoda	500	IHI	1986	IV	IHI	1986	H
Chugoku Elec.	Shin-Onoda	500	IHI	1987	IV	IHI	1987	H
Hokkaido Elec.	Tomato	350 ⁱ	BHK	1980	IV	BHK	1980	L
Hokkaido Elec.	Tomato	600	MHI	1984	IV	-	-	-
Joban Joint	Nakoso ^j	600	MHI	1983	IV	MHI	1983	H
Joban Joint	Nakoso ^j	600	MHI	1983	IV	IHI	1983	H
Kyushu Elec.	Omura	156	MHI	1982	IV	-	-	-
Kyushu Elec.	Minato	156	MMMC	1983	III	MHI	1983	H
Kyushu Elec.	Matsuura	700	-	1987	-	MHI	1987	H
Sakata Joint	Sakata	350	MHI	1984	IV	MHI	1984	H
Sakata Joint	Sakata	350	MHI	1987	IV	MHI	1987	H
Shikoku Elec.	Saijo	156	KHI	1983	VI	MHI	1983	H
Shikoku Elec.	Saijo	250	KHI	1984	VI	IHI	1984	H
Tohoku Elec.	Sendai	175 x 2	BHK	1983	VI	BHK	1983	H
Tokyo Elec.	Yokosuka ^k	265	MHI	1985	IV	MHI	1984	H
Tokyo Elec.	Yokosuka ^k	265	BHK	1985	IV	BHK	1985	H
Toyama Joint	Toyama	200 x 2	Chiyoda	1984	V	BHK	1984	H

^aMitsui Miike Machinery Co.

^bIshikawajima-Harima Heavy Industries.

^cBabcock Hitachi K. K.

^dKawasaki Heavy Industries.

^eLow-dust system.

^fSubjecting 75% of gas to FGD.

^gMitsubishi Heavy Industries.

^hHigh-dust system.

ⁱSubjecting 50% of gas to FGD and 25% of gas to SCR.

^jBurns both oil and coal.

^kBurns a coal/oil mixture.

TABLE 2-7. CLASSIFICATION OF WET LIME/LIMESTONE PROCESSES

Type	By-Product	Absorbent	Additive	Prescrubber ^a	Sulfuric Acid
I	Sludge	Ca(OH)_2	None	None	None
II	Gypsum	CaCO_3	Sodium	None	None
III	Gypsum	CaCO_3	Sodium	None	Yes
IV	Gypsum	CaCO_3	None	Yes	Yes
V	Gypsum	CaCO_3	None	Yes	None
VI	Gypsum	Ca(OH)_2	MgO^b	Yes	None

^aWith a separate liquor loop.

^bNo need when lime contains over 1% MgO .

using two scrubbers in series. These plants are owned by EPDC. In addition, MMC has completed new plants with a single scrubber for Kyushu Electric and Mitsui Coal.

Type IV FGD systems have a prescrubber with a separate liquor loop and use sulfuric acid for pH adjustment. This system was developed by Mitsubishi Heavy Industries (MHI), and has been applied to many oil-fired and coal-fired boilers.

Most new FGD plants for coal-fired boilers use a Type IV FGD system, as shown in Table 2-6. The prescrubber in this system has the following functions: 1) It removes fly ash in order to increase the purity of by-product gypsum. 2) It removes chloride in the flue gas; chloride tends to increase corrosion and to lower SO_2 removal efficiency. 3) It cools the gas to prevent scaling and to protect scrubber linings. 4) It removes fluoride in the flue gas which tends to lower SO_2 removal efficiency in combination with aluminum derived from fly ash.

It has been shown that fluorine and aluminum combine to form various types of polymerized complex ions which prevent chemical reactions (7). It

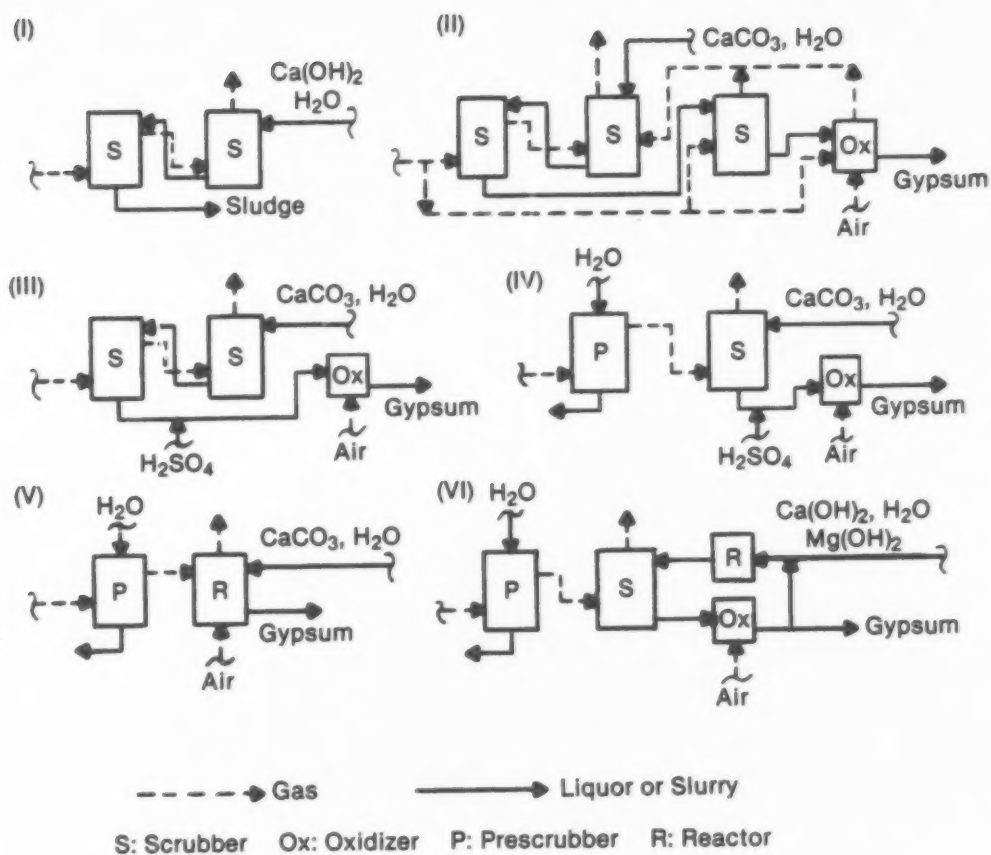
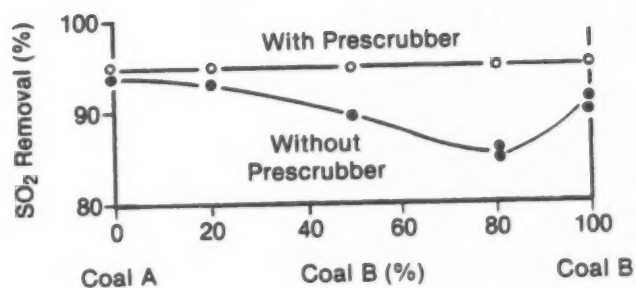


Figure 2-15. Wet lime/limestone processes.

appears likely that the complex ions cover the surface of the calcium carbonate, thus lowering its reactivity. These ions also cover the surface of the calcium sulfite and delay the oxidation process. The combined effect of fluoride and aluminum is especially noticeable with certain type of coals or a blend of coals (Figure 2-16).

The prescrubber removes both fluorine and fly ash (aluminum) to prevent this combined effect. The behavior of the sodium may be due to the formation of an inert compound such as Na_3AlF_6 . A prescrubber may not be needed for a coal containing little fluoride or when a reaction promoter, such as sodium, magnesium or organic acid, is added. Without the prescrubber, a small amount of fly ash goes into the by-product gypsum but does not affect its suitability for use in cement or wallboard. Somewhat offsetting its beneficial effects, the prescrubber raises the FGD plant cost by 3-7% and increases the amount of wastewater produced.

Recently, two new FGD processes have been applied to coal-fired boilers: the Chiyoda Jet Bubbling Reactor Process (Type V) and the Kawasaki magnesium-gypsum process (Type VI). Both processes attain perfect oxidation without using sulfuric acid and produce salable gypsum (refer back to Tables 2-6 and 2-7).



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Figure 2-16. Coal blending and SO₂ removal efficiency of the limestone-gypsum process (Babcock Hitachi).

SECTION 3

NEW FGD PLANTS

3.1 FEATURES OF NEW FGD PLANTS FOR COAL-FIRED UTILITY BOILERS

The operation parameters of nine FGD plants for coal-fired utility boilers are shown in Tables 3-1 and 3-2. Eight plants use the wet limestone-gypsum process while Shikoku Electric's Saijo plant uses lime containing about 1% magnesia to produce by-product gypsum. The scrubbers remove 90-98% of the SO_2 and 70-94% of the particulates.

In these new FGD systems, the flue gas leaves the scrubber at around 50°C and is heated to about 100°C by a Ljungstrom gas-gas heater (GGH). The one exception is Joban's Nakosa plant where the gas is heated to 120°C by a GGH and a steam-gas heater. In the GGH, a small amount of the gas (2-4%) leaks, resulting in slightly decreased SO_2 and particulate removal efficiencies. Only EPDC's Takehara Station which uses the system D gas-gas heater shown in Figure 2-8 has almost no gas leakage. All FGD plants completed between 1979 and 1984 use a rotating gas-gas heater, while the older plants use oil-firing to heat the gas. Non-leak type gas-gas heaters will be used at Tokyo Electric's Yokosuka plants now under construction.

FGD plants for coal-fired boilers completed before 1978 use a venturi or perforated plate scrubber. These scrubbers have a high dust removal efficiency but cause a large pressure drop, resulting in high power consumption (2.4-2.6%). Spray tower absorbers have become popular since 1981 because of the low pressure drop and lower power consumption (2.0-2.2%)

TABLE 3-1. FGD PLANTS BUILT FOR COAL-FIRED BOILERS (1979-83)

Owner/ Station	Chugoku/ Shimonoseki	Hokkaido/ Tomato	EPDC/ Matsushima	Tohoku/ Sendai
Boiler Capacity, MW	175	350 ^a	500 x 2 ^b	175 x 2
Boiler start-up year	1962	1980	1981	1962
FGD start-up year	1979	1980	1981	1983
FGD type	IV	IV	IV	IV
FGD constructor	MHI	BHK	IHI, BHK	BHK
Gas treated, 1,000 Nm ³ /h	610	610	1,300 x 2	549 x 2
Inlet SO ₂ , ppm	730	230	1,000	360
Inlet dust, mg/Nm ³	830	25	300	55
1st scrubber (prescrubber)	Spray	Venturi	Spray	Spray
pH	1	1	1	1
L/G, liter/Nm ³	3	2.2	3	2.5
2nd scrubber (scrubber)	Packed	pp ^c	Spray	Spray
pH	5.5	-	-	-
L/G, liter/Nm ³	14	5	16	12
Outlet SO ₂ , ppm	45	10	60	27
Outlet dust, mg/Nm ³	50	5	30	6
SO ₂ removal, % ^d	94	96	94	93
Dust removal, % ^d	94	80	90	87
Gas reheating	GCH ^e	GCH	GCH	GCH
Pressure drop, mm H ₂ O	240 ^f	450 ^g	150 ^h	390 ^g
Wastewater, t/h	15	2.2	15 x 2	10 x 2
Power consumption, % ⁱ	2.2	1.7	2.2	2.1
Investment cost, yen/kW ^j	34,000	-	-	40,000

^aSubjecting 50% of gas to FGD.^bSubjecting 75% of gas to FGD.^cPerforated plate.^dSlightly lower after GCH.^eGas-gas heater.^fBy scrubbers and mist eliminators.^gBy scrubbers, mist eliminators and GCH.^hBy scrubbers.ⁱPercent of power generated by the boiler.^jIncluding gas reheating system and civil engineering.

TABLE 3-2. FGD PLANTS FOR COAL-FIRED BOILERS (1983-84)

Owner/ Station	Joban/ Nakoso ^a	EPDC/ Takehara	Kyushu/ Minato	Shikoku/ Saijo	Toyama/ Toyama
Boiler capacity, MW	600x2	700	156	156	200x2
Boiler start-up year	1983	1983	1983	1983	1984
FGD type	IV	III	III	VI	V
FGD constructor	MHI	IHI	MMC	KHI	Chiyoda
Gas treated, 1,000 Nm ³ /h	1,860x2	2,200	560	560	730x2
Inlet SO ₂ , ppm	1,000	700	1,520	700-800	630
Inlet dust, mg/Nm ³	150	30-150	100	30-100	100
1st scrubber (prescrubber)	spray	spray	none	spray	venturi
pH	1	4.5-5	-	1	1
L/G, liter/Nm ³	2	5	-	2	1
2nd scrubber (scrubber)	packed	spray	spray	spray	JBR ^b
pH	5.5	5.5-6	5.5	5.5-6	4
L/G, liter/Nm ³	20	15	16	4	-
Outlet SO ₂ , ppm	40	30-35	10-30	15-20	63
Outlet dust, mg/Nm ³	45	10-20	10-20	10-20	10-20
SO ₂ removal, % ^c	96	95	98-99	98	90
Dust removal, % ^c	70	70-80	80-90	70-80	80-90
Gas reheating	GGH & SGH ^d	GGH	GGH	GGH	GGH
Pressure drop, mm H ₂ O ^e	340	350	280	480	630
Wastewater, t/h	13	25	5	5	4
Power consumption, % ^f	1.8	2.1	2.0	1.9	1.7
Investment cost, Yen/kW ^g	21,000	20,000	24,000	20,000	19,000

^aBurning oil and coal (90:10)^bJet bubbling reactor^cSlightly lower after GGH^dSteam-gas heater (to 120°C)^eBy scrubbers, mist eliminators, and GGH^fPercent of power generated by the boiler^gIncluding gas reheating system and civil engineering

associated with them. Mitsubishi Heavy Industries (MHI) continues to use a plastic grid-packed low pressure-drop tower with power consumption of 1.8-2.0 percent. The power consumption is particularly low (1.7%) for Hokkaido Electric's Tomato-Atsuma plant due to the low inlet SO_2 concentration and the use of an improved perforated plate scrubber (Figure 2-4). Toyama Power's Toyama plant has the lowest power consumption (1.7%) due to the use of a Jet Bubbling Reactor without slurry recirculation pumps (Section 3-5).

The investment cost of most of the plants completed since 1983 is about 20,000 yen/kW, much lower than that of plants completed between 1977 and 1979. The cost reduction is due mainly to technology improvements which allow the plant to be simplified without reducing its reliability. All of the plants have been operated with over 99% reliability.

The three FGD plants constructed between 1979 and 1981 were described in SO_2 Abatement for Coal-Fired Boilers in Japan (EPA-600/7-83-028). The six plants completed in late 1983 and 1984 are described below.

3.2 EPDC'S TAKEHARA NO. 3 PLANT, (IHI)

3.2.1 Process

The FGD plant for the No. 3 coal-fired boiler (700 MW) at EPDC's Takehara Station was constructed by Ishikawajima Harima Heavy Industries (IHI) using the Type III wet limestone gypsum process (Figure 2-15). It began commercial operation in 1983. At Takehara, the train made up of two Chemico spray scrubbers may be one of the largest in the world. Hot side ESPs and SCR reactors are installed upstream of FGD for efficient dust removal with various coals.

The Type III FGD process was chosen for two main reasons. First, with Type III, very efficient hot side ESPs can be installed to keep the dust content low at the FGD inlet. This results in less fly ash contamination of the by-product gypsum, even though the fly ash is not removed by a prescrubber

with a separate liquor loop, as it would be with a Type IV system. Secondly, if Type IV were used, the prescrubber would produce a small amount of sludge which could not be conveniently disposed of at Takehara.

3.2.2 Plant Flowsheet

A flowsheet of the Takehara plant is shown in Figure 3-1. Flue gas from the boiler ($2,400,000 \text{ Nm}^3/\text{h}$) is treated by two trains of hot ESP and SCR reactors, and then treated in two gas-gas heaters in parallel. An FGD fan is located between the GGH and scrubber and a scavenging fan is added to prevent leakage of the untreated gas into the cleaned gas. The flue gas is then introduced into the first scrubber with a two-stage spray, and from there into the main scrubber with a six-stage spray. The cleaned gas is passed through mist eliminators and then is heated by the gas-gas heater.

Limestone slurry is fed into the main scrubber and a portion of the circulating slurry in the main scrubber is fed into the first scrubber or prescrubber. The calcium sulfite slurry leaving the first scrubber is acidified with a small amount of sulfuric acid and then oxidized by air bubbles in oxidizers. The resultant gypsum is separated by a thickener and a centrifuge.

3.2.3 Specifications

The design specifications of the system at Takehara are as follows:

Inlet gas volume: $2,400,000 \text{ m}^3$ (normal)/h(wet)

Inlet gas temp.: 100°C (min. 70°C)

SO_x concentration

Desulfurization system inlet: 1,000 ppm

Desulfurization system outlet: 100 ppm or less

Dust Load

Desulfurization system inlet: 200 mg/m^3 (normal)

Desulfurization system outlet: 25 mg/m^3 or less (normal)

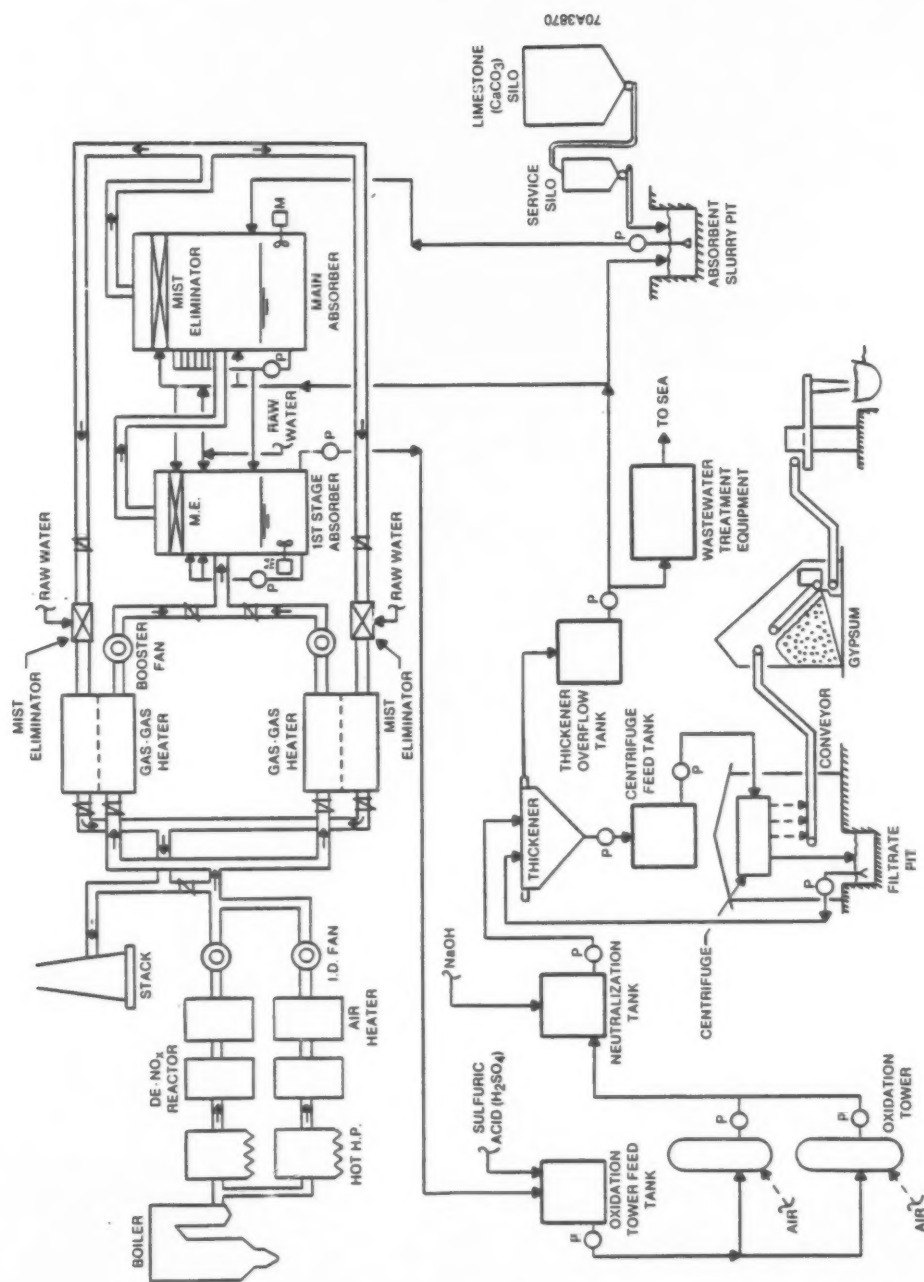


Figure 3-1. Takehara FGD system.

Gypsum

Free moisture content: 10% (weight) or less
Purity: 95% (weight) or more

The dimensions and specifications of the major equipment at the Takehara plant are shown in Table 3-3.

TABLE 3-3. MAJOR EQUIPMENT AT THE TAKEHARA PLANT

(a) <u>Towers</u>			
	Diameter (m)	Height (m)	Remarks
Prescrubber	18.3	27.5	Spray type
Absorber	18.3	42.0	Spray type
Oxidation tower	7.2	13.0	2 units
(b) <u>Pumps</u>			
	Capacity (m ³ /min)	Unit	Spare Unit
For prescrubber	56	6	1
For absorber	53	18	6
(c) <u>Other</u>			
Boost-up fans	Fan capacity: 32,900 m ³ /min Motor output: 5,100 kW No. of units: 2		
Gas-gas heaters	Heat transfer area: 29,790 m ² /unit No. of units: 2		
Centrifuges			
Type	Model 60 (basket type)		
No. of units	12		

3.2.4 Performance

The results of actual performance tests versus design data for the Takehara FGD plant are shown in Table 3-4. Plant operation has been successful with no serious problems. After 2,100 hours of operation, the plant was shut down for inspection. A small amount of soft deposit-containing water was found in the duct between the GGH and the first scrubber, but this did not hinder plant operation. Some scale about 0.5-1 mm thick was found at the top of the mist eliminator in the first scrubber. The mist eliminator washing method has since been changed to prevent this scale build-up. Power consumption has been relatively low, averaging 2.0-2.1% of the power generated by the boiler.

3.2.5 Economics and Plant Evaluation

Both plant costs and annualized operation costs at Takehara are quite low, 20,000 yen/kW and 1.5 yen/kWh respectively, and SO₂ removal efficiency is high. A small amount of sodium is added to neutralize the gypsum slurry. The sodium goes into the scrubber and oxidizer system and may work to promote the absorption and oxidation process. Plant operation has been successful. The plant cost may be further lowered if the first scrubber can be eliminated so that there is just one tower system.

3.3 JOBAN JOINT POWER'S NAKOSO PLANT, (MHI)

3.3.1 Introduction

Joban Joint Power Co. constructed two 600 MW boilers (No. 8 and No. 9) at the Nakoso Station. The boilers, which began operating in 1983, are designed to burn both coal and oil, with an extensive flue gas treatment system including SCR, ESP, and FGD. Since the local residents were concerned about possible pollution caused by extensive coal-firing, the boiler began operation by burning coal and oil at a ratio of 10:90. The flue gas leaving the wet scrubber was reheated to 120°C using a Ljungstrom gas-gas heater and a

TABLE 3-4. RESULTS OF PERFORMANCE TESTS AT THE TAKEHARA FGD PLANT

Parameters	Design	Test Operation
Boiler load, MW	MCR	700
Inlet gas, m ³ /h (normal)	2,400,000	2,085,000
Dust load		
Inlet, mg/m ³ (normal)	200	70
Outlet, mg/m ³ (normal)	<25	20
SO ₂ content		
Inlet, ppm	1,000	505
Outlet, ppm	<100	34
Removal efficiency, %	>90	93.3
By-product Gypsum		
Free moisture, weight %	<10.0	6.65
Purity, weight %	>95.0	98.1
Mist load		
Outlet of prescrubber, g/m ³ (normal)	0.9	0.38
Outlet of absorber, g/m ³ (normal)	0.9	0.60
Inlet of gas-gas heater, g/m ³ (normal)	0.15	0.14
Calcium carbonate, t/h (purity 98.9%)	9.5	4.7
Sulfuric acid, t/h (purity 98.0%)	0.45	0.06
Caustic soda, t/h (conc. 25.0%)	0.49	0.04
By-product gypsum, (t/h)	18.5	8.2
Industrial water, (t/h)	112.2	115.0
Electric power consumption, MW	17.4	14.0

steam-gas heater. Since gas cleaning proved satisfactory, Joban Joint Power increased its coal/oil ratio to 30:70 in April 1984. A larger amount of coal may be burned in the future.

For Unit No. 8, MHI supplied the boiler, SCR, ESP, and FGD plants for Unit No. 9; IHI supplied the boiler and SCR plant, and MHI the ESP and FGD plants.

3.3.2 FGD Plant and Operation

The flue gas cleaning system at the Nakoso Station is shown in Figure 3-2. Each boiler has two cleaning system trains with a Ljungstrom type gas-gas heater and a steam-gas heater. The flowsheet of the FGD plant is similar to the flowsheet in Figure 2-3. Table 3-5 gives the sizes of the major FGD equipment at the plant. The small prescrubber has a separate liquor loop to remove impurities in the flue gas, such as fly ash, fluoride, and chloride. The standard MHI scrubber has a low-pressure drop plastic grid packing. Flue gas passes downward through both the prescrubber and the scrubber along with the liquor and slurry, resulting in a low pressure drop of 120 mm H₂O through the prescrubber and scrubber.

An example of the operational results when the coal: oil ratio was 10:90 is shown in Table 3-2. About 96% of the SO₂ and 70% of the dust was removed by the prescrubber and scrubber. Table 3-6 shows the planned operating parameters for firing coal and oil in a ratio of 30:70. Actual operating results have been close to the planned values. In addition, the FGD plants have experienced no problems since startup.

The Nakoso FGD plants have the following noteworthy features.

- (1) They have a low power consumption of only 1.8%. This is especially low for a limestone gypsum FGD plant with a high SO₂ removal efficiency (Tables 3-1 and 3-2). Low power consumption is a result of the low pressure drop due to the cocurrent flow; use of an axial flow FGD fan whose blade angle can be changed according to the flue gas volume; and other devices.
- (2) They only experience about 1% gas leakage in the Ljungstrom gas-gas heater. A high SO₂ removal efficiency is attained at the stack even with the FGD fan at the "A" position (Figure 2-9).

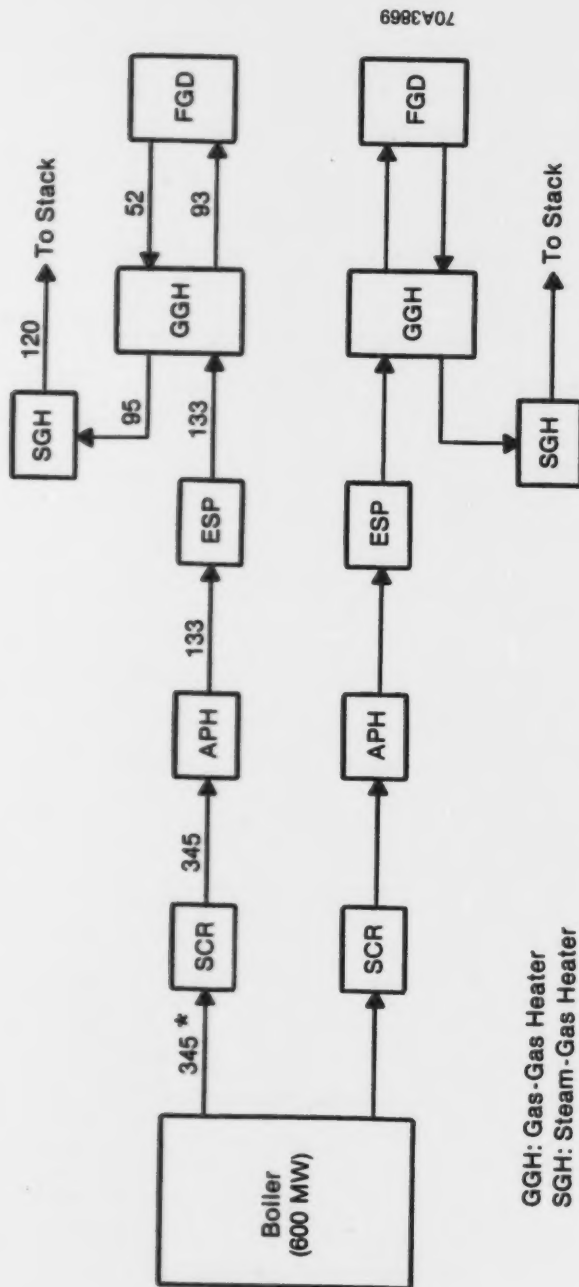


Figure 3-2. Boiler and flue gas treatment system at Joban Joint Power's Nakoso station.

TABLE 3-5. MAJOR FGD EQUIPMENT FOR ONE TRAIN (300 MW)

Equipment	Number	Size (m) or Capacity
Prescrubber	1	4.0 x 6.4 x 6.7 (height)
Scrubber	1	12.7 x 6.7 x 36.5 (height)
Pump (prescrubber)	2 + 1 ^a	1250 m ³ /h
Pump (scrubber)	5	4250 m ³ /h
Oxidizer	1	3.9 (diameter) x 17.2 (height)
Centrifuge	6 + 1 ^a	

^aSpare.TABLE 3-6. OPERATION OF FGD PLANTS AT NAKOSO STATION
(Projected figures for a coal:oil ratio of 30:70)

Flue gas, Nm ³ /h	1,860,000
Inlet SO ₂ , ppm	Below 930
Inlet dust, mg/Nm ³	120
Prescrubber L/G, l/Nm ³	2
Scrubber L/G, l/Nm ³	20
pH	5.5
Gas-gas heater leakage, %	1
Stack inlet SO ₂ , ppm	Below 88
Stack inlet dust, mg/Nm ³	Below 50
Wastewater, m ³ /h	13.5
Power consumption, %	1.83

The investment cost of the Nakoso plant is about 21,000 yen/kW, which is considered to be fairly low.

3.3.3 Evaluation

The low power consumption of the Nakoso Station is an attractive feature. As shown in Tables 3-1 and 3-2, the power consumption of the Nakoso plants may be the lowest of the limestone gypsum process plants except for Hokkaido Electric's Tomato plant and Toyama Joint Power's Toyama plant. The Tomato plant treats a flue gas with an unusually low SO₂ concentration and the Toyama plant uses a special type of scrubber (Section 3.5).

Table 3-7 compares the Nakoso No. 8 and the Takehara No. 3 FGD plants. Both treat flue gas with about the same concentrations of SO₂ and dust with about equal efficiency. The Takehara plant does not have a prescrubber with a separate liquor loop but has a prescrubber as the first absorber. The prescrubber at the Nakoso plant is considerably smaller than the one at the Takehara plant and Nakoso's main scrubber is also considerably smaller in m³/MW. Both the L/G ratio and the pressure drop are nearly equal at the two plants, while the power consumption is lower at Nakoso.

The fact that the Nakoso plant has a smaller scrubber but achieves about the same efficiency level as a larger scrubber may be due to the use of an efficient semi-venturi spray tower for the prescrubber and a packed tower for the main scrubber. By comparison, the Takehara plant uses simple spray towers. The relatively low pressure drop associated with the use of the semi-venturi and packed tower may be the result of good design and the cocurrent flow in the scrubbers. The lower power consumption may be due mainly to the smaller amount of power needed by the slurry recirculation pumps; the height of the tower is lower at the Nakoso plant. In addition, the Takehara spray tower may require a certain type of pump head to produce fine sprays.

TABLE 3-7. COMPARISON OF NAKOSO AND TAKEHARA FGD PLANTS

	Nakoso (No. 8)	Takehara (No. 3)
Capacity, MW	600	700
Inlet SO ₂ , ppm	900	700
SO ₂ removal by scrubbers, %	96	95
Inlet dust, mg/Nm ³	120 - 150	30 - 150
Dust removal by scrubbers, %	70	70 - 80
Prescrubber volume, m ³	172 x 2	7,729
m ³ /MW	0.57	10.3
Prescrubber height, m	6.7	27.5
Main scrubber volume, m ³	3,106 x 2	11,041
m ³ /MW	10.4	15.8
Main scrubber height	36.5	42.0
L/G, 1/Nm ³ (both scrubbers total)	22	20
Pressure drop, mm H ₂ O	340	350
Power consumption, %	1.8	2.1
Wastewater, m ³ /h	15	25

The advantages of the Nakoso plant may partially be due to the fuel it uses -- a small amount of coal with a larger amount of oil. If coal alone is used, the amount of wastewater from the prescrubber will increase because of the need to remove a larger amount of chloride, but the operation of the main scrubber will not change significantly.

3.4 NEW MITSUI MIIKE (MMC) FGD PLANTS

3.4.1 Development Status

Mitsui Miike Engineering Co., (formerly Mitsui Miike Machinery Co. (MMC)) has constructed several lime/limestone FGD plants for coal-fired boilers in Japan, Germany, and The Netherlands using Chemico (GE) type scrubbers. MMC recently simplified the process and constructed two plants for coal-fired boilers, both of which began operation in 1983. One is for Kyushu Electric's Minato Station (156 MW) and the other is for Mitsui Coal Mining Co. at Omuta (47.5 MW). MMC's process is a simplified version of a Type III FGD system (Figure 2-15).

3.4.2 Process

A flowsheet of the FGD process at Kyushu Electric's Minato plant is shown in Figure 3-3. The flowsheet of Mitsui Coal's Miike plant is the same except that at the Miike plant, the fan for FGD is placed between the FGD outlet and the GGH, as shown in Figure 2-9, C. The plants have a single spray tower absorber where about 95% of the SO_2 is absorbed by a limestone slurry, which leaves the scrubber at a pH of 5.5. After a small amount of sulfuric acid is added to lower its pH, the slurry is introduced into an oxidizing tower for complete oxidation of the calcium sulfite into gypsum. The gypsum slurry is treated with a thickener and a centrifuge to produce a salable gypsum by-product containing less than 10% moisture with over 95% purity.

The circulating liquor contains a small amount of sodium which serves as a catalyst or promoter for SO_2 absorption and oxidation. A portion of the liquor is purged after wastewater treatment to prevent the accumulation of impurities such as chloride. Sodium sulfate or hydroxide is added for make-up sodium.

3.4.3 Performance

The operation data of the two MMC FGD plants are shown in Table 3-8. A high SO_2 removal efficiency--97-98% at Minato and over 98% at Miike -- has

TABLE 3-8. OPERATIONAL PARAMETERS FOR TWO MMC FGD PLANTS

Parameter	Kyushu Electric's Minato Plant (156 MW)		Mitsui Coal's Omuta Plant (47.5 MW)	
	Design	Actual	Design	Actual
Inlet SO ₂ , ppm	1,500	1,100-1,200	3,200	2,700
Inlet dust, mg/Nm ³	100	20-50	400	<100
L/G, l/Nm ³	16	16	17	17
Absorbent pH	5.5-6.0	5.5	6.0-6.2	5.6
CaCO ₃ stoichiometry	1.05	<1.05	1.10	1.08
Wastewater, m ³ /h	5	5	6	6
Gypsum purity, %	95	96-98	95	98
Total pressure drop, mm H ₂ O	350	280	500	450
GGH outlet temperature, °C	104	106	85	87
GGH leakage, %	5.5	2.5	5	<0.5
Scrubber outlet				
SO ₂ , ppm	70	10-30	118 ^a	<50 ^a
Dust, mg/Nm ³	34.5	10-20	28 ^a	<10 ^a
Mist, mg/Nm ³	150	130	150	<20
Power consumption, %		2.0		2.5-3
Plant cost, yen/kW		24,000		45,000

^a At stack inlet

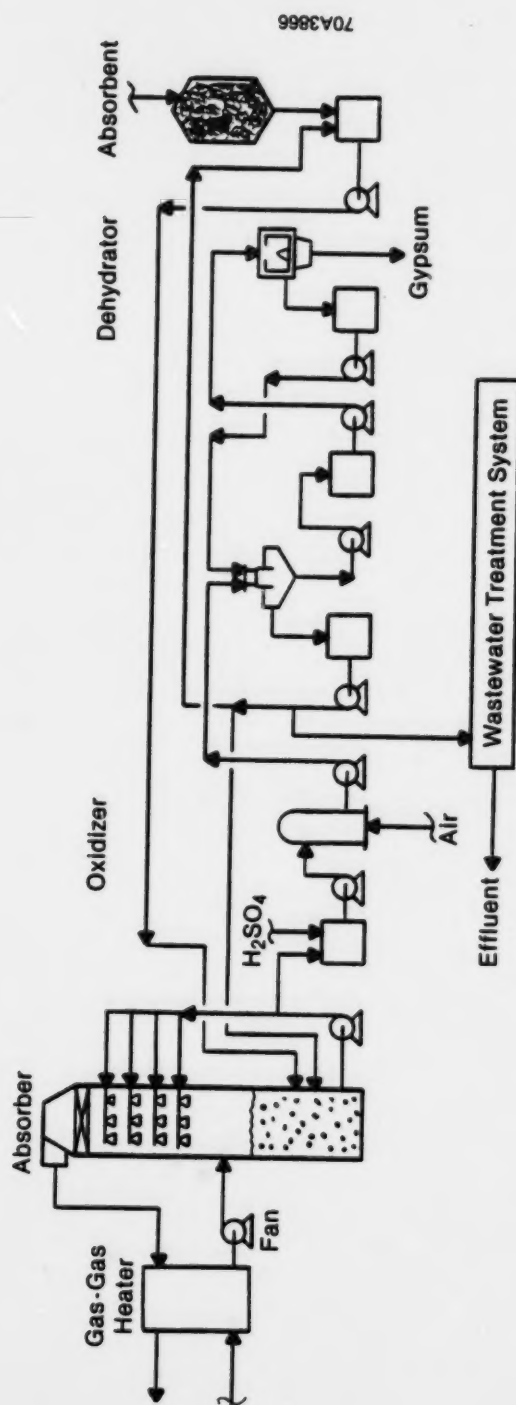


Figure 3-3. Kyushu Electric Co. - Minato FGD plant (limestone-gypsum process).

been obtained using a single scrubber system. At Minato, the FGD fan is placed at the "A" position (Figure 2-9) and about 2.5% of the untreated gas leaks into the cleaned gas. At Miike, the fan is placed at the "C" position to maintain as low an SO_2 concentration at the stack as possible. A fan at the "C" position is a little more costly than a fan at the "A" position because corrosion-resistant materials must be used at "C." At Minato, the "A" position is used because the inlet SO_2 concentration is quite low and the regulations can be met with the fan in that position. Both of the plants have been operated without significant problems.

3.4.4 Economics and Evaluation

The FGD plant cost for the Minato plant was 24,000 yen/kW, fairly low for a system that was retrofit to a 156 MW boiler. The Miike plant cost was 45,000 yen/kW. This is still fairly low for a small FGD plant including a GGH and wastewater treatment system. The simplicity of FGD plants with a single scrubber contributes to the low investment cost. It appears that sodium addition works efficiently to increase SO_2 absorption and sulfite oxidation, so that over 97% SO_2 removal efficiency can be attained with a single scrubber.

Tests at the Minato plant reveal that over 95% oxidation can be attained in the scrubber and that salable gypsum may be obtained without an oxidizing tower. The high oxidation ratio may be due to the relatively low inlet SO_2 concentration and the effect of sodium. Elimination of the oxidizing tower and sulfuric acid addition makes the process more economical. The simplified process is similar to a process being developed in the U.S. MMC's simplified process and the Chiyoda Jet Bubbling Reactor process described in Section 3.5 may be the simplest and most economical FGD processes for the production of by-product gypsum.

3.5 TOYAMA JOINT POWER'S TOYAMA PLANT (Chiyoda JBR Process)

3.5.1 Introduction

Chiyoda has constructed two commercial FGD plants for industrial boilers. Both plants use the CT 121 process and the Jet Bubbling Reactor, and have been operated successfully. In 1984, Chiyoda completed construction of two FGD units for Toyama Joint Power's 200 MW coal-fired boilers. The boilers originally were designed as 250 MW oil-fired boilers and have been modified to burn either coal at 200MW or oil at 250MW. The design basis of the FGD plants is shown in Table 3-9.

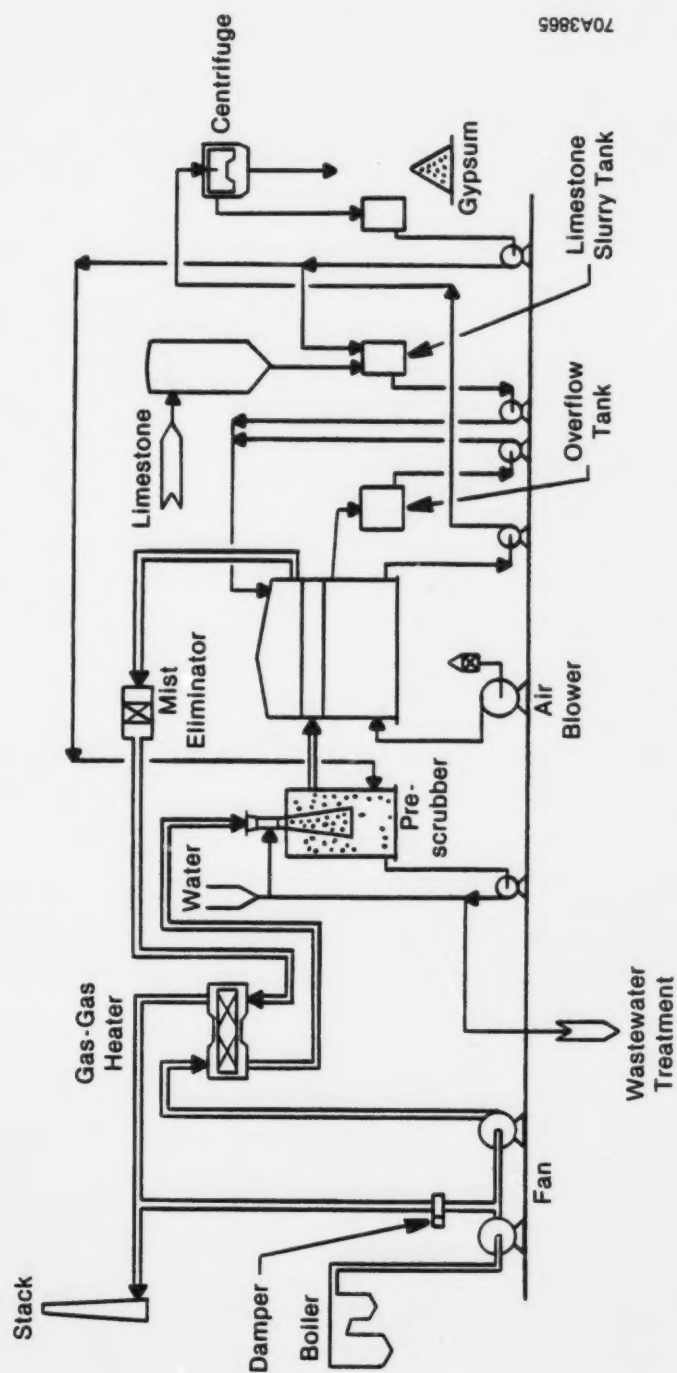
TABLE 3-9. DESIGN BASIS OF TOYAMA FGD PLANTS

	Coal	Oil
Sulfur in Fuel, %	1.0	1.2
Gas Volume, Nm ³ /h	705,000	760,000
Capacity, MW	200	250
SO ₂ removal, %	Over 80 ^a	Over 80 ^a
CaCO ₃ , t/day	47	42
Gypsum, t/day	91	82

^aOver 86% at scrubber outlet

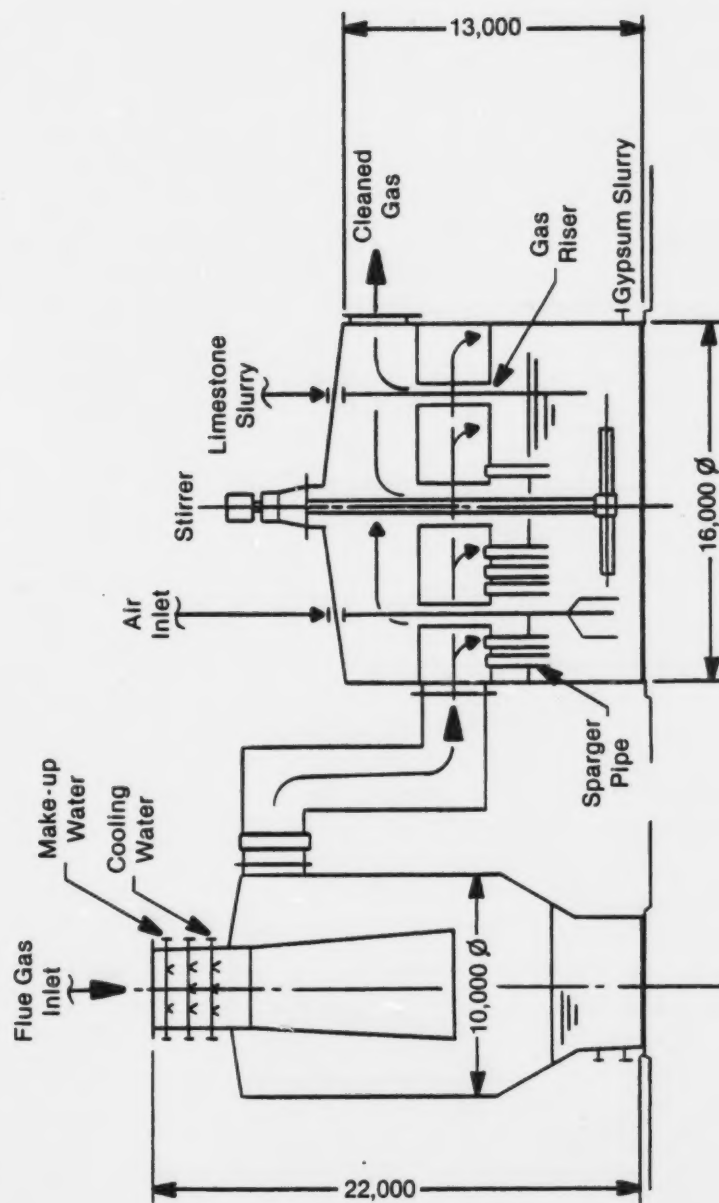
3.5.2 FGD Plants (8)

A flowsheet of the Toyama plant is shown in Figure 3-4. Table 3-10 gives the dimensions of the major equipment at the plant. The plant has a Jet Bubbling Reactor with a prescrubber in a separate liquor loop (Figure 3-5). In the reactor, flue gas is injected into a limestone-gypsum slurry through many submerged sparger pipes, while limestone slurry is fed into the reactor and air for oxidation is blown into the slurry. Limestone reacts with SO₂ and



70A3865

Figure 3-4. Flowsheet for the Chiyoda CT-121 JBR Process (Toyama Plant).



70A3864

Figure 3-5. Prescrubber and Jet Bubbling Reactor (in mm).

O₂ to form gypsum, which is discharged from the reactor. Neither slurry recirculation pumps nor an oxidation tower is required. The space requirement for the system is small--12,000 m² for the two plants, including a wastewater treatment system. If necessary, this requirement can be reduced to 90,000 m².

TABLE 3-10. MAJOR EQUIPMENT FOR A CHIYODA CT-121 JBR FGD UNIT

	Number	Type	Dimensions or Capacity
Prescrubber, m (diameter/height)	1	Semi-venturi	10/22
JBR scrubber, m (diameter/height)	1	Jet Bubbling	16/13
Over-flow tank, m (diameter/height)	1	Cylinder	4.3/5
Limestone silo, m (diameter/height)	1	Cylinder	8/8.3
Gas-gas heater	1	Ljungstrom	29 VIX-2150
Mist eliminator	1	Euroform (3 stages)	
Flue gas fan, Nm ³ /h	1	Turbo	780,000
Centrifuge, t/h (each)	2+1 (spare)	Basket	1.7
Pump for prescrubber, m ³ /h	1		500

Typical operation parameters for the Toyama plant are shown in Table 3-2. The scrubber removes 90% of SO₂ and 93% of dust and consumes a relatively small amount of power -- 1.7%. If necessary, the SO₂ removal efficiency can be raised by increasing the sparger submersion depth or by raising the slurry pH as shown in Figures 3-6 and 3-7. During actual operation tests with oil-firing (250 MW, 785,000 Nm³/h), 90 percent SO₂ removal efficiency was attained with 1.5% power consumption.

The plant has an improved Ljungstrom gas-gas heater for reheating the gas from 43 to 103°C. A scavenging fan is used to minimize flue gas leakage. The

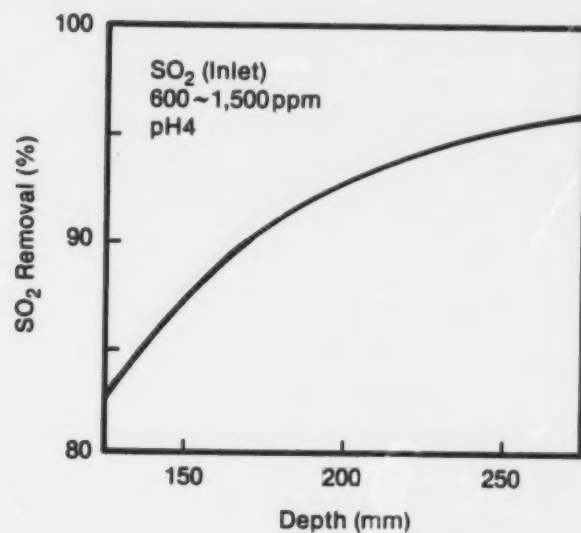


Figure 3-6. Submersion depth of gas sparger pipes vs. SO₂ removal efficiency.

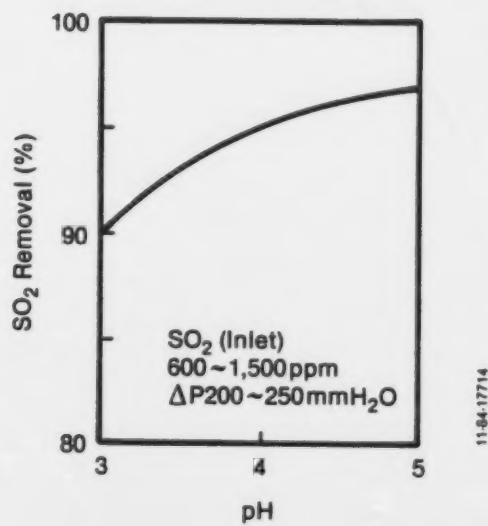


Figure 3-7. pH vs. SO₂ removal efficiency.

heating elements are in two, rather than the conventional three, sections. The length of a section is 1,075 mm, possibly the longest ever used in Japan. Soot blowing works better when this type of element is used to prevent dust deposition. The Euroform mist eliminator used was designed to reduce mists (maximum 5,000 mg/Nm³) to less than 150 mg/Nm³.

3.5.3 Evaluation

The operation parameters and investment costs of four new gypsum-producing FGD plants are compared in Table 3-2. The Chiyoda process has both the lowest investment cost (yen/kW) and power consumption (%). The low power consumption is due to the lack of slurry recirculating pumps for the scrubber.

The power consumption of the Chiyoda process may be a little higher when over 95% SO₂ is removed, as is the case with the other FGD plants in Table 3-2. On the other hand, power consumption might be slightly lower if the plant were constructed for coal-firing only. However, when both coal and oil are fired, coal-firing has a higher power consumption (1.7%), than oil (1.5%). This is due partially to the larger gas volume/kW with coal and partly to the fact that the plant is designed to treat 785,000 Nm³/h of flue gas for 250 MW oil-firing. The fan and other equipment can treat more than 730,000 Nm³/h for 200 MW coal firing. Power consumption of the plant will be further reduced through the use of an axial fan.

The CT-121 process has a relatively high pressure drop. Without lowering the SO₂ removal efficiency, the pressure drop may be reduced by raising the slurry pH and reducing the submersion depth of the gas sparger pipes. Chiyoda's tests have shown that no scaling occurs when the slurry has a pH less than 4.5. When the pH is raised to 4.5, the pressure drop will decrease, but the by-product gypsum probably will contain a small amount of unreacted limestone. Usually, Japanese users of gypsum want the limestone content below 1%.

Chiyoda's JBR process has the following advantages: 1) ease of operation; 2) high reliability; 3) lower labor requirements; 4) low space requirements; 5) lower investment and operation costs; 6) production of high quality gypsum by-product; and 7) formation of only a small amount of COD (mainly dithionate) that requires wastewater treatment.

The major disadvantage of the CT-121 process was the limited reactor size. However, Chiyoda currently can construct a single large reactor with the capacity to treat flue gas from a 700 MW coal-fired boiler.

3.6 TOHOKU ELECTRIC'S SENDAI PLANT (BABCOCK HITACHI)

3.6.1 Overview

Tohoku Electric's Sendai Power Station has three 175 MW boilers, each constructed between 1959 and 1962. These radiant-heat, single-drum type boilers have natural circulation and are designed for coal burning. Until recently the boilers burned low sulfur oil with a small amount of coal.

SCR and FGD plants have been installed for the No. 2 and No. 3 boilers which burn only coal. Babcock Hitachi constructed the plants; one (for the No. 3 boiler) began operation in February 1983, while the other (No. 2) started up in May 1983. The boilers consume 1,660 t/day each of coal imported from Australia, South Africa, Canada, etc., containing 0.2-1.2% S and 0.8-1.7% N.

The No. 1 boiler has neither FGD nor SCR; it uses 670 t/day of low-sulfur coal with 580 kl/day of low-sulfur oil. The regulations for the three Sendai station boilers are shown in Table 3-11.

The flue gas cleaning system for the No. 2 and 3 boilers is shown in Figure 3-8. This high-dust system has a cold-side ESP downstream of the SCR reactor.

TABLE 3-11. REGULATIONS AND AGREEMENTS FOR THE SENDAI STATION BOILERS

Parameter	Central Government Regulations		Agreements with Local Governments
SO _x , Nm ³ /h	No. 1	308	Total 868
	No. 2	280	
	No. 3	280	
NO _x , ppm at 6% O ₂	480		No. 1 300
			No. 2 260
			No. 3 260
Particulates, mg/Nm ³	400		150

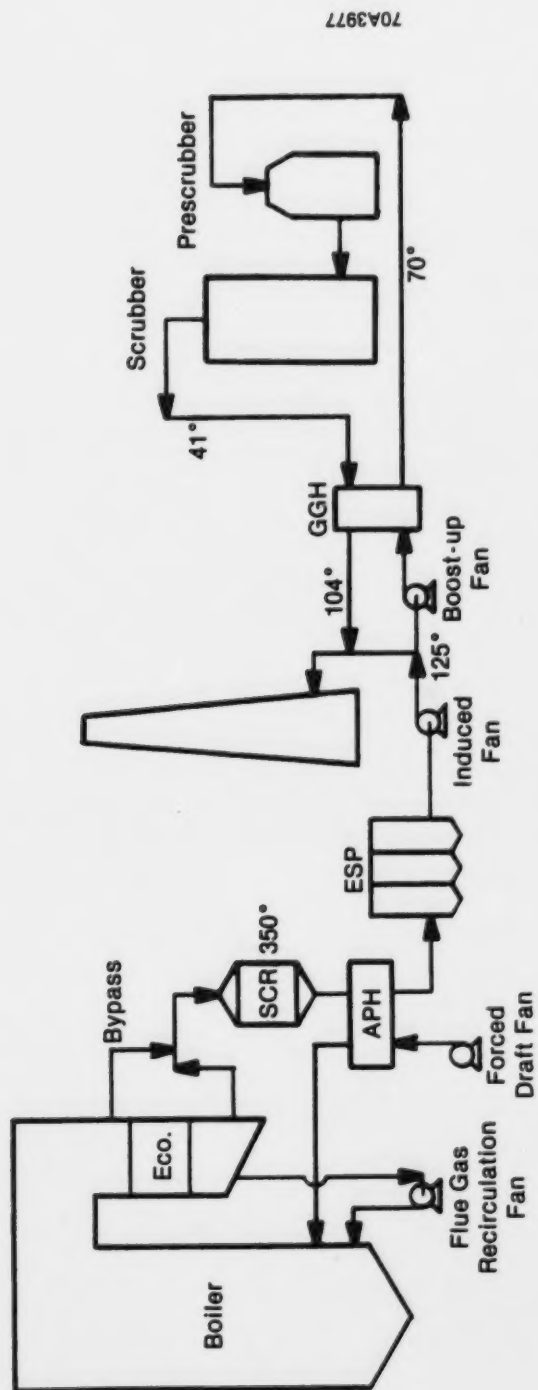
3.6.2 ESP and GGH

The ESP normally reduces particulates in the flue gas from 15-20 g/Nm³ to about 0.1 g/Nm³ at 125°C. This represents a removal efficiency of 99.3-99.5%. After it leaves the ESP, the gas is sent to a Ljungstrom gas-gas heater and cooled to about 70°C. In the GGH, the gas leaving the scrubber at 41°C is heated to 104°C. The leakage in the GGH is 4%. The GGH heating elements are coated with enamel and are subjected to soot blowing four times a day. Steam at 15 kg/cm² is used at a rate of 3,000 kg/h.

The GGH has been operated without dust plugging problems since plant startup. Low inlet dust content (usually about 0.15 g/Nm³) and efficient soot blowing seem to prevent dust deposition.

3.6.3 FGD Plant

The flue gas passing through the GGH is introduced into a prescrubber with a separate liquor loop and treated with water at an L/G ratio of about 2.2 l/Nm³. The cooled gas is then sent to a spray tower absorber and treated with a limestone slurry at an L/G ratio of about 11 l/Nm³. The design and an



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Figure 3-8. Flue gas cleaning system of the No. 2 and No. 3 boilers at the Sendai Station.

example of the actual performance of the FGD system are shown in Table 3-12. Outlet SO₂ and dust concentrations are very low because of the low inlet concentrations and the high removal efficiency of the FGD plant.

TABLE 3-12. PERFORMANCE OF AN FGD PLANT FOR A 175 MW COAL-FIRED UNIT

Items	Design	Actual
Gas Flow Rate, Nm ³ /h	675,000	640,000
Inlet SO ₂ Conc., ppm	1,010	330
Outlet SO ₂ Conc., ppm	152	39
SO ₂ Removal Efficiency, %	85	88.1
Inlet Dust Load, mg/Nm ³	632	126
Outlet Dust Load, mg/Nm ³	150	41
Gas Temperature		
Gas Inlet, °C	150	125
Gas Outlet, °C	104	105
By-product Gypsum		
Surface Moisture, %	<11	5.2
Purity, %	95	99

The pressure drop of the gas across the FGD system, including the GGH, is about 390 mm H₂O. The FGD plant requires 27 t/h of water and produces 10 t/h of wastewater. The wastewater treatment system is similar to those used for other new FGD units for coal-fired boilers (see Figure 2-10). Because the ammonia content of the wastewater is low (30-50 mg/l), no ammonia removal treatment is needed. After it is treated, most of the wastewater is sprayed on the coal in a storage yard.

The FGD plant has a pH control system that uses sulfuric acid and also an air bubbling oxidizer. Both of these systems are almost unnecessary because of the high oxidation ratio of calcium sulfite to gypsum in the scrubber due to the low inlet SO_2 concentration (high O_2/SO_2 ratio of the gas). The by-product gypsum contains 6-8% moisture with little ammonia (0.002%) and is sold for wallboard production and as a cement setting retarder.

The investment cost of the Sendai FGD plants was 14 billion yen for 350 MW. This included the cost of the GGH and excluded civil engineering costs.

3.6.4 Evaluation

The flue gas cleaning system at Sendai is similar to that at Chugoku Electric's Shimonoseki Power Station. The Shimonoseki plant was constructed by MHI and has been in operation since 1980. Unlike the Shimonoseki plant, the Sendai flue gas cleaning system has not experienced any problems with dust deposition in its FGD gas-gas heater. At Shimonoseki, the gas-gas heater must be washed with water twice a year in order to remove the dust deposited within it.

The difference in dust deposition between the two systems may be a result of differing SO_3 contents in flue gases. The SO_3 content of the gas at Sendai may be much lower for the following reasons: 1) the SO_2 content is much lower than at Shimonoseki (330 ppm vs. 500-820 ppm) and 2) the SCR plant at Sendai was completed in 1983 while the Shimonoseki plant was completed in 1980. In recent years, SCR catalysts have been improved to oxidize less SO_2 to SO_3 . The lower dust content of the gas at Sendai (below 150 mg/Nm^3 vs. over 400 mg/Nm^3 at Shimonoseki) may also have some effect on dust deposition. (See Section 2.6.1). The fly ash at both plants may contain about 0.01% NH_3 . It appears that the small amount of ammonia compounds formed on the surface of the fly ash has little effect upon dust deposition in the gas-gas heater.

The FGD system at Sendai has been operated without problems since startup. However, this system does have a high investment cost of 40,000 yen/kW. The high cost may be due to the small size of the units that were retrofitted to the existing boilers.

3.7 SHIKOKU ELECTRIC'S SAIJO PLANT (KHI Magnesium Gypsum Process)

3.7.1 Development Status

Kawasaki Heavy Industries (KHI) developed the magnesium-gypsum process and has constructed two units for oil-fired boilers. Recently KHI constructed a unit to treat one-third of the flue gas from a 700 MW coal-fired boiler at the Mehrum Station, West Germany. The unit began operation in 1981. KHI also constructed FGD units for two coal-fired boilers (156 and 250 MW) at Shikoku Electric's Saijo Station. These units began operation in 1983 and 1984, respectively.

3.7.2 Process Description

A basic flowsheet of the KHI process is shown in Figure 3-9. Although the Mehrum plant does not have a gas-gas heater, it does have an oil-fired gas reheater. The Saijo plants each have a gas-gas heater and a prescrubber upstream of the absorbing tower. The prescrubber has a separate liquor loop and removes fly ash in order to increase the quality of the by-product gypsum.

Sulfur dioxide is absorbed by a slurry containing $\text{Ca}(\text{OH})_2$, $\text{Mg}(\text{OH})_2$ and gypsum. The hydroxides in the slurry react with the SO_2 to form MgSO_3 , CaSO_3 , $\text{Mg}(\text{HSO}_3)_2$, and $\text{Ca}(\text{HSO}_3)_2$. The slurry is sent to an oxidizing tower where the sulfites and bisulfites are oxidized into sulfates and bisulfates. The slurry is treated with a thickener and centrifuge to separate the gypsum by-product. The mother liquor is treated with lime to produce a slurry containing $\text{Ca}(\text{OH})_2$, $\text{Mg}(\text{OH})_2$ and gypsum which is sent to the absorber. The relationship of the slurry pH and L/G ratio to the SO_2 removal efficiency is shown in Figure 3-10. Magnesium increases SO_2 removal efficiency and prevents gypsum scaling.

Specifications for the KHI FGD units for coal-fired boilers are shown in Tables 3-13 and 3-14. The Mehrum unit is designed to treat one-half of the gas from the boiler ($1,224,000 \text{ Nm}^3/\text{h}$) but actually has been treating one-third

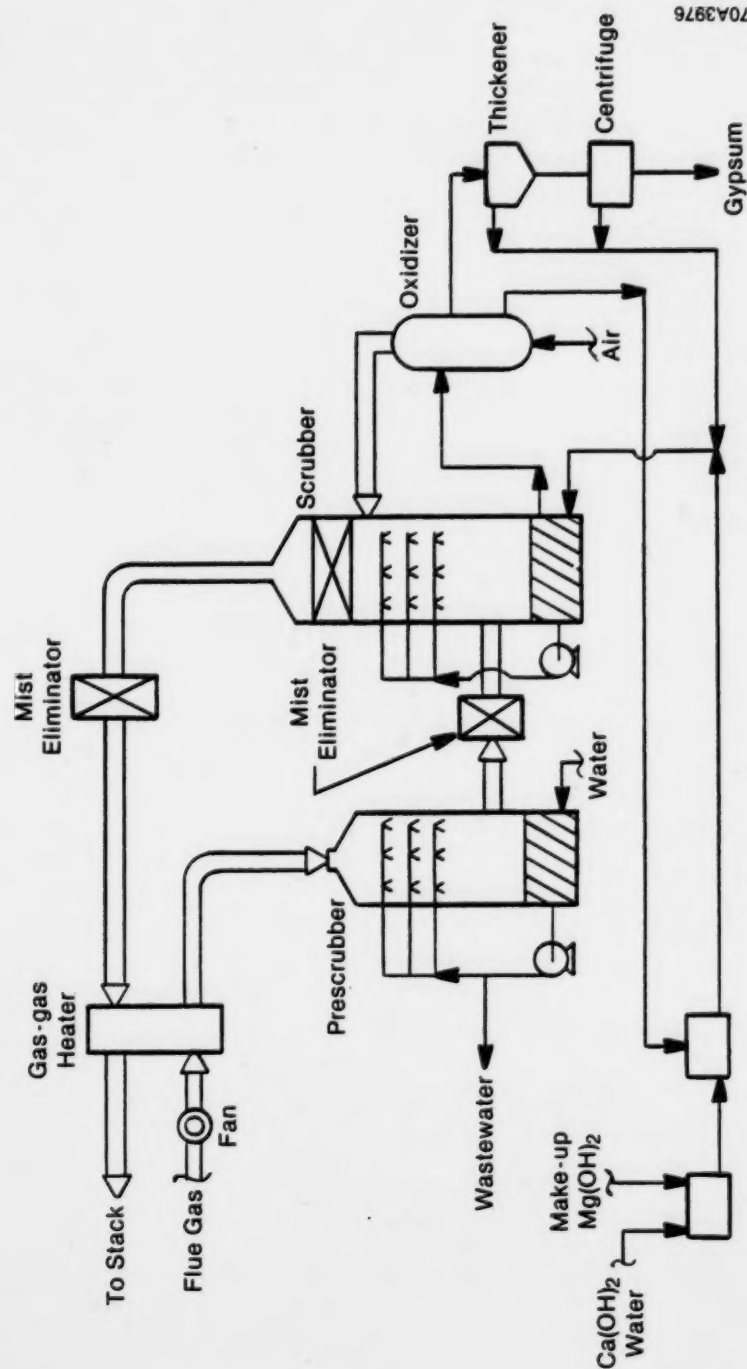
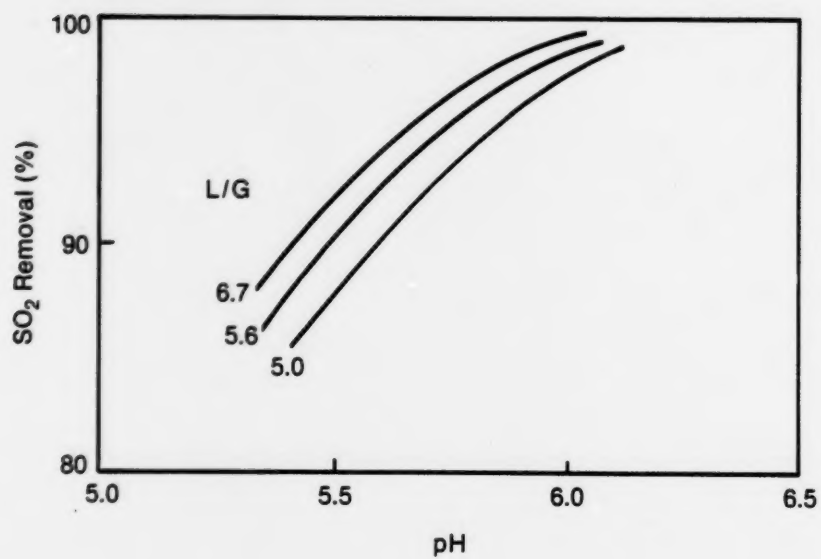


Figure 3-9. Flowsheet for the Saijo plant.



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Figure 3-10. Absorbent pH, L/G ratio, and SO₂ removal efficiency for the KHI Process.

TABLE 3-13. FGD SPECIFICATIONS FOR COAL-FIRED BOILERS (3)

Item	Mehrum No. 3	Saijo No. 1	Saijo No. 2
Start-up Date	Sept. 1981	Apr. 1983	Oct. 1983
Unit Output, MW	700	156	250
Treating Ratio	1/3	1/1	1/1
Treating Gas Volume, Nm ³ /h	816,000	557,000	858,000
Inlet Concentration (design value)			
SO ₂ , ppm	851 (wet)	820 (dry)	914 (dry)
HCl, ppm	95 (wet)	34 (dry)	36 (dry)
HF, ppm	12 (wet)	74 (dry)	79 (dry)
Particulate, mg/Nm ³	100 (wet)	100 (dry)	100 (dry)
Outlet Concentration (design value)			
SO ₂ , ppm	110 (Absorber)	82 (Stack)	91 (Stack)
Particulate, mg/Nm ³	<100 (Absorber)	30 (Stack)	30 (Stack)
SO ₂ removal efficiency, %	87 (Absorber)	90 (Overall)	90 (Overall)
Particulate efficiency, %	-	70 (Overall)	70 (Overall)
Alkali	CaO	Ca(OH) ₂	Ca(OH) ₂
Magnesium	MgSO ₄ ·H ₂ O	Mg(OH) ₂	Mg(OH) ₂
By-product	Gypsum	Gypsum	Gypsum
CaSO ₄ ·2H ₂ O (dry)	4.6 t/h	3.1 t/h	5.0 t/h
Effluent Liquor, m ³ /h	20	7	12
Sulfuric Acid	None	None	None
Reheating Method	After burning	GGH (2 trains)	GGH (2 trains)
Reheating Temp., °C	95	98	98

of the gas (816,000 Nm³/h). The unit has no prescrubber and purges 20 t/h of wastewater from the scrubber system to maintain a chloride concentration

TABLE 3-14. MAJOR EQUIPMENT OF THE KHI FGD PLANTS(3)

	Mehrum	Saijo No. 1	Saijo No. 2
Prescrubber type, size, m	None	Spray 9.80 x 21	Spray 12.10 x 23
Absorber type size, m	Spray tower 14.50 x 32.5	Spray tower 9.80 x 31	Spray tower 12.10 x 33
Mist eliminator, type	Chevron	Chevron	Chevron
Recirculation pump			
For prescrubber, capacity	- -	3 + (1)* 680 m ³ /h	3 + (1) 1025 m ³ /h
For absorber, capacity	3 + (1) 2900 m ³ /h	4 + (1) 1220 m ³ /h	4 + (1) 1890 m ³ /h
Oxidizer, number	1	2 (series)	2 (series)
Centrifuge Type	4 + 1 Basket	4 + 1 Basket	5 + 1 Basket

*The number in parentheses refers to standby equipment.

below 10,000 ppm in the scrubber liquor. The by-product gypsum contains about 100 ppm chloride after being water-washed in the centrifuge, and is used for wallboard.

The flue gas at Saijo contains less chloride but much more fluoride than the gas at Mehrum. The prescrubber in the Saijo units removes fluoride and chloride in order to increase the SO₂ removal efficiency. The units have a

water-softening system to reduce wastewater; the purge liquor from the scrubber is treated with sodium hydroxide and carbonate to precipitate magnesium and calcium, and the supernatant is sent to the prescrubber.

3.7.3 System Performance

The operation data for the Saijo plants are shown in Table 3-15. The performance of the two plants are similar except that the No. 2 plant uses a smaller (4 vs. 5) L/G ratio than the No. 1 plant and attains a higher SO₂ removal efficiency (98% vs. 95%). KHI feels that this is due to the higher sulfite concentration in the scrubber liquor of the No. 2 plant. The flue gas from the No. 1 boiler is rich in O₂ (8%) and about 90% of the sulfites are oxidized into sulfates in the scrubber absorber. Flue gas from the No. 2 boiler contains 5.5% O₂ and experiences a 50% oxidation rate in the scrubber. Since magnesium sulfite is an important SO₂ absorbent, a higher sulfite concentration results in a higher SO₂ removal efficiency.

Magnesium is not added to the Saijo FGD systems because the lime used there contains 0.8-1.0% MgO. This amount of MgO allows the desired Mg⁺² concentration of 0.5-1.0% to be maintained in the scrubber liquor.

Operation of the Saijo plant has been successful with no significant problems. The by-product gypsum is of a good quality and is used both for wallboard and cement.

3.7.4 Economics and Evaluation

The investment cost of the Saijo plant is 20,000 yen/kW including the cost of the GGH and civil engineering. This price is fairly low for this relatively small plant. The small size of the scrubber and slurry recirculation pumps and the absence of a pH controller for sulfuric acid addition may contribute to the low cost. On the other hand, lime is considerably more costly than limestone. Overall, it is difficult to say whether or not the KHI process is less costly than the limestone-gypsum process.

TABLE 3-15. OPERATIONAL PARAMETERS OF THE SAIJO FGD PLANT

	No. 1	No. 2
Boiler capacity, MW	156	250
Gas treated, Nm ³ /h	561,000	873,000
Inlet SO ₂ , ppm	700 - 1,000	700 - 1,000
Inlet dust, mg/Nm ³	30 - 100	30 - 100
Inlet O ₂ , %	8.0	5.5
Prescrubber, type	Spray	Spray
L/G, l/Nm ³	2	2
pH	1	1
Main scrubber		
Type	Spray	Spray
pH	5.5 - 6	5.5 - 6
L/G, l/Nm ³	5	4
Outlet SO ₂ , ppm	35 - 50	15 - 20
Outlet dust, mg/Nm ³	10 - 20	10 - 20
SO ₂ removal, %	95	98
Dust removal, %	70 - 80	70 - 80
Wastewater, m ³ /h	5	8
Pressure drop, mm H ₂ O	480	360
Power consumption	1.9	1.8
Plant cost, yen/kW	20,000	20,000

KHI estimates that the cost of its FGD plant is about 6% lower when the prescrubber is not installed. Elimination of the prescrubber does not seem to produce any adverse effect when the flue gas has a composition similar to that of the Saijo plant. In that situation, the inlet dust concentration is not high and there is no substantial negative effect on the gypsum quality. The adverse effects of fluoride and chloride may be lessened by the effect of the magnesium. One reason why Japanese utility companies prefer to produce high purity gypsum is that in the future there may be an oversupply of gypsum. This would make it impossible to sell low grade gypsum, thereby creating a disposal problem.

3.8 NEW FGD PLANTS FOR VARIOUS GASES (Kureha, Nippon Steel, Dowa, and Chiyoda)

3.8.1 Overview

Table 3-16 shows major new FGD plants for treating gases other than those produced by coal combustion. Because of the recent economic downturn, only a few new FGD plants have been constructed, other than those for coal-fired boilers, since 1982. Existing plants already have sufficient countermeasures for SO₂ emission controls. Most of the new FGD plants are either for boilers burning inexpensive low-grade, high-sulfur oil which require a high SO₂ removal efficiency of over 97% (Toyo Rubber's Sendai plant and Mitsubishi Petrochemical's Yokkaichi plant) or for waste gases from a steel producer or a smelter with a low SO₂ concentration (Nippon Steel's Sakai plant and Mitsui Metal's Miike plant). One of the reasons for the installation of an FGD plant for the latter is to reduce the SO₂ levy (Table 1-9).

All of the new FGD plants are quite economical with low investment costs and low power consumption. They generally use an inexpensive absorbent such as limestone or industrial slag.

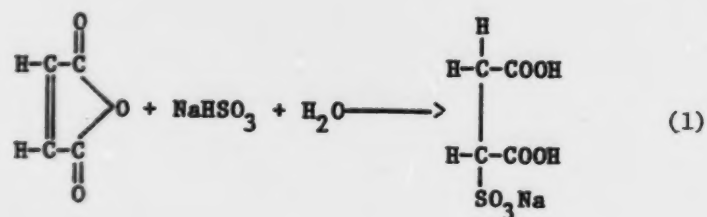
TABLE 3-16. MAJOR NEW FGD PLANTS FOR VARIOUS GASES

Owner/ Plant Location	Toyo Rubber/ Sendai	Nippon Steel/ Sakai	Mitsui Metal/ Miike	Mitsubishi Petrochem./ Yokkaichi
Gas Source	Oil-fired boiler	Sintering machine	H ₂ SO ₄ plant	Oil-fired boiler
Gas volume, Nm ³ /h	40,000	550,000x2	141,000	250,000
MW equivalent	13	180x2	45	80
FGD plant constructor	Kureha	Nippon Steel	Dow	Chiyoda
FGD start-up year	1982	1982	1983	1983
Absorbent	CaCO ₃ +SS ^a	Converter slag	Al ₂ (SO ₄) ₃ CaCO ₃	CaCO ₃
Inlet SO ₂ , ppm	1,500	100-300	250	1,500
Inlet dust, mg/Nm ³	150	10-30	10	180
Prescrubber (1st scrubber)				
Type	none	venturi	none	venturi
pH	-	5-6	-	1
L/G, l/Nm ³	-	4-8	-	1
Scrubber				
Type	PP ^b	venturi	packed	JBR ^c
pH	5.5	7	3.5	3.5
L/G, l/Nm ³	2.7	4	-	-
Outlet SO ₂ , ppm	5	2-3	5	35
Outlet dust, mg/Nm ³	70-80	Below 10	1	35
SO ₂ removal, %	99.7	98-99	98	98
Dust removal, %	50	50	90	78
Pressure drop, mm H ₂ O	200	400	150	600
Power consumption, %	1.4	1.4-1.6	-	1.3
Wastewater, m ³ /h	0	1.5-2	-	1.5
Investment cost				
Yen/Nm ³	6,300	3,180	-	6,120
Yen/kW equivalent	19,000	10,000	-	18,000

^aSodium Sulfo succinate^bPerforated Plate^cJet Bubbling Reactor

3.8.2 Kureha Organic Acid-Limestone Gypsum Process

Kureha Chemical Industries (KCI) has extensively studied the use of organic compound additives to the lime/limestone gypsum process and has found sulfosuccinate to work well. When anhydrous maleic acid is added to a scrubber liquor containing a small amount of sodium, the acid readily reacts to form sodium sulfosuccinate as shown in Equation 1. Sodium sulfosuccinate increases the SO₂ removal efficiency of the lime/limestone slurry.



Anydrous + Sodium bisulfite present —————> Sodium
maleic acid in scrubber liquor sulfosuccinate

Since May 1982, the Toyo Rubber plant (Kureha acetate-lime process, 40,000 Nm³/hr, oil-fired boiler) has used maleic acid instead of acetic acid and since November 1982, limestone has been used instead of lime (Figure 3-11). Because the sulfosuccinate is not volatile, the acetic acid recovery section in the scrubber is not needed and the scrubber has been simplified, resulting in a smaller pressure drop. The pH of the slurry in the reactor (scrubber discharge) is about 5. The use of limestone has not required any modifications in the reactor.

The Toyo plant has been operated successfully using limestone and maleic acid. Due to the reduced production of rubber, the oil-fired boiler produces 25,000 Nm³/h of flue gas (8 MW equivalent). The power consumption of the FGD plant is 90 kW, about 1.1% of the power to be generated.

The concentration of sulfosuccinate in the scrubber liquor is less than 1%. Both maleic acid and sulfosuccinate are used in cosmetics and other

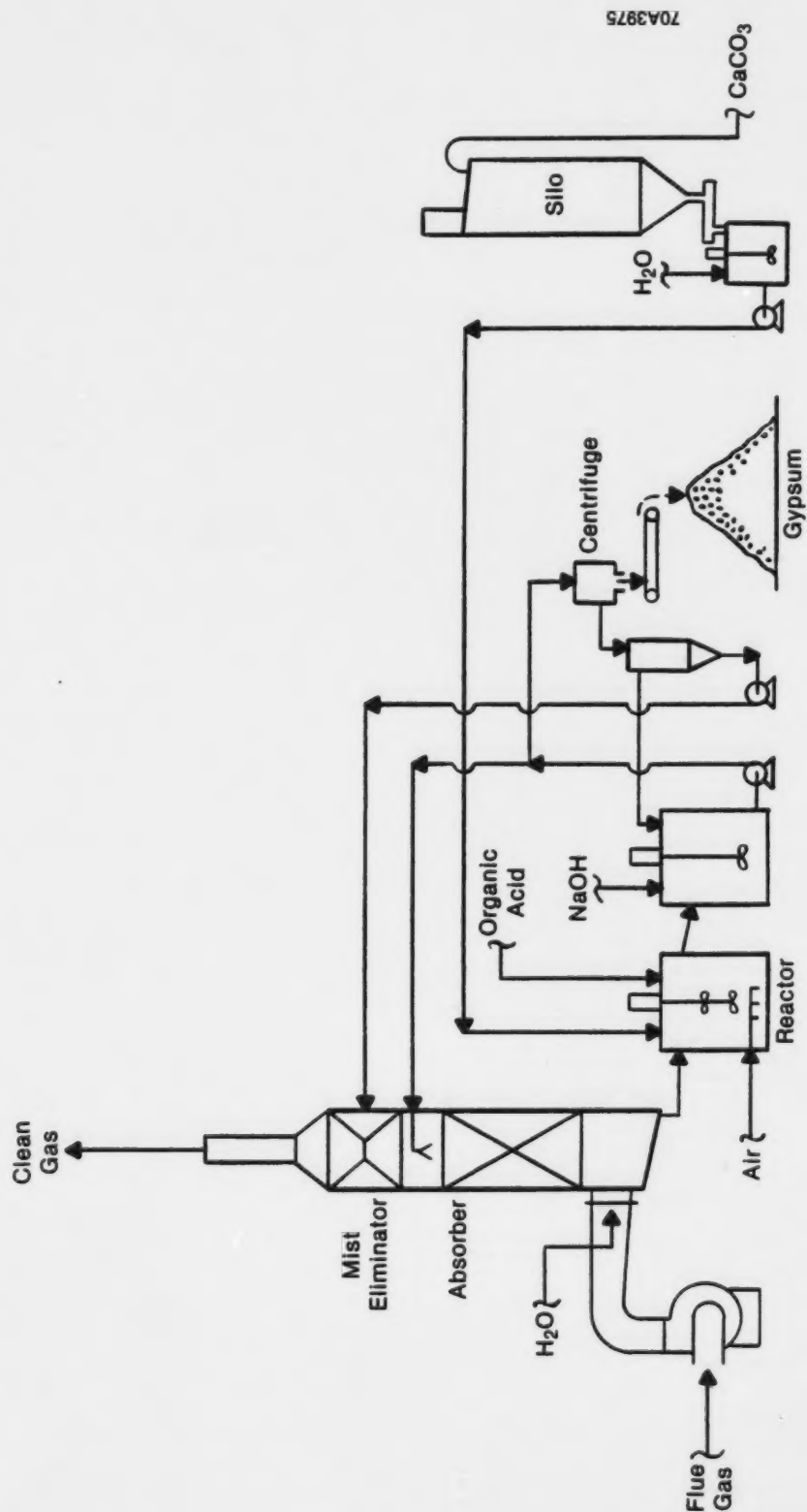


Figure 3-11. Flowsheet of the Kureha organic acid limestone gypsum process.

products and are not hazardous. The cost of maleic acid is about 280 yen/kg, about twice that of acetic acid.

The sulfosuccinate, as well as acetate and adipate, is decomposed gradually through FGD. The decomposition ratios of those compounds are nearly proportional to the oxidation of calcium sulfite to gypsum. However, as shown in Table 3-17, the sulfosuccinate decomposition ratio is smaller than those for acetate and adipate.

TABLE 3-17. DECOMPOSITION RATIO OF ORGANIC ACIDS
(Acid decomposed/ CaSO_3 mol $\times$ 100, at pH 6.8,
 Cl^- 0.47%) (Initial concentration 98 mol/kg
as RCOO^-)

	Acetate	Sulfosuccinate	Adipate
Decomposition ratio	3.1	2.1	2.8

Overall, the sulfosuccinate-limestone process is better than the acetate-lime process because the former uses limestone and a simple scrubber with less pressure drop. The sulfosuccinate-limestone process may be similar to the limestone-adipic acid process used in the U.S. However, the decomposition product of the sulfosuccinate process has no odor as does the product from adipic acid. Kureha feels that sulfosuccinate is more effective than adipate as an additive. Further studies are needed to compare the effects of sulfosuccinate and adipate.

3.8.3 Converter Slag Scrubbing

Nippon Steel Co., the largest steel producer in Japan, has developed an FGD process using converter slag slurry as the absorbent to by-produce disposal gypsum. Converter slag, which is produced in large amounts in steel plants, contains about 40% CaO , 35% FeO and Fe_2O_3 , 3% MgO , and about 16% SiO_2 , and has little use. The first converter slag FGD plant was completed in 1976 at the Wakamatsu Plant and had a capacity of treating $1,000,000 \text{ Nm}^3/\text{h}$ of flue

gas from an iron-ore sintering machine. Nippon Steel has improved the process and constructed two units at Sakai, each with a capacity of treating 550,000 Nm³/h of flue gas from an iron-ore sintering machine. These units went into operation in 1982 (Table 3-16).

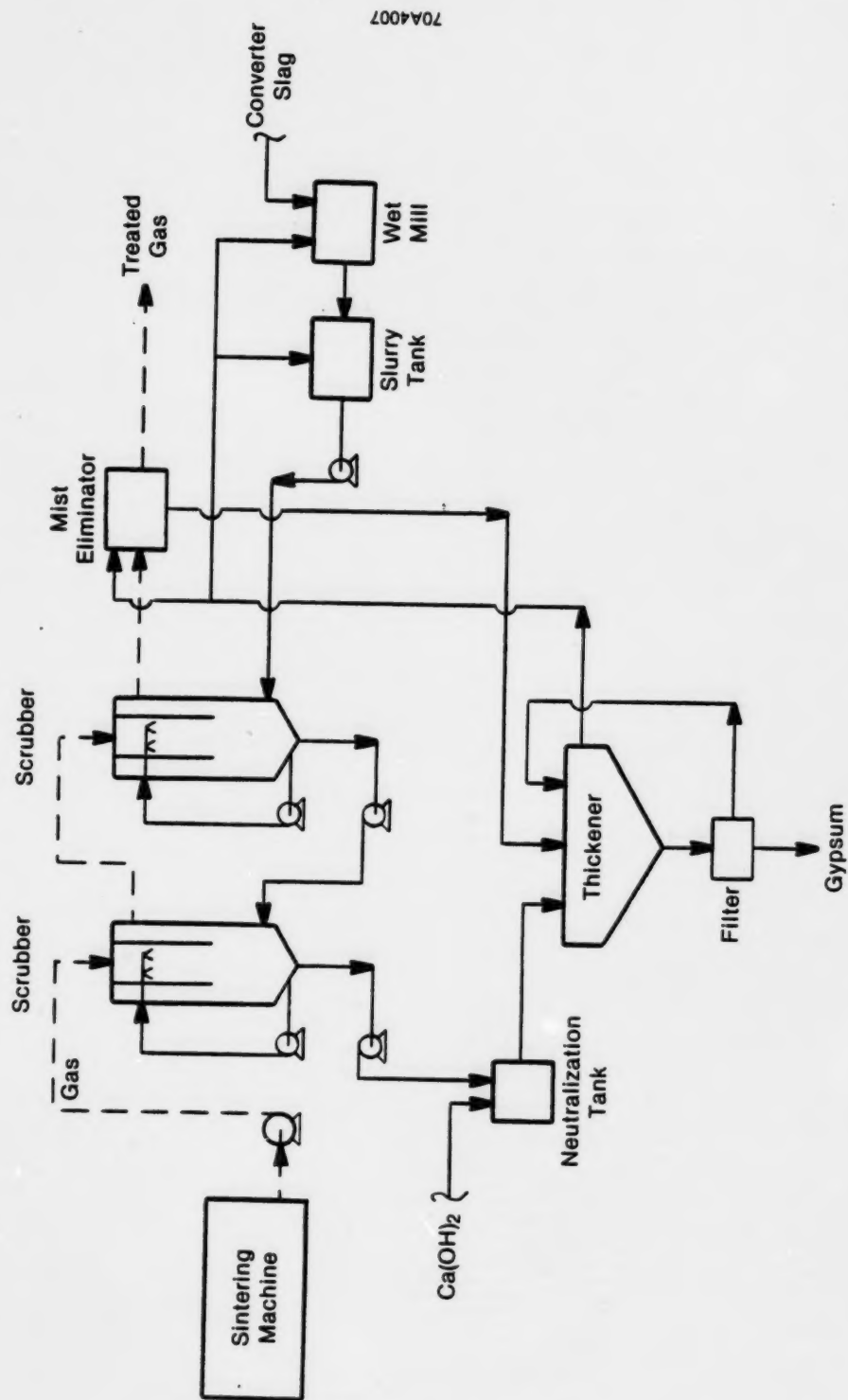
The flue gas is treated by a slurry containing powdered converter slag to remove more than 98% of the SO₂ (Figure 3-12). In the scrubber, nearly all of the calcium sulfite is oxidized into gypsum for the following reasons: 1) the flue gas contains low concentrations of SO₂ (100-300 ppm) and a large amount of O₂ (about 15%) and 2) the converter slag contains significant amounts of FeO and Fe₂O₃, which act as catalysts for the oxidation reaction. The gypsum by-product contains about 40% impurities and is discarded.

The plant cost, 10,000 yen/kW, is unusually low. The low cost may be due in part to the fact that the FGD plant was constructed by the steel company itself. The process may become more economical if the prescrubber can be eliminated, and may be suitable to steel plants which produce large amounts of converter slag.

3.8.4 Dowa Aluminum Sulfate-Gypsum Process

The Dowa process uses aluminum sulfate liquor (pH 3.5) as the absorbent and limestone as the precipitant. The process flowsheet is shown in Figure 3-13. The process is simple, easy to operate, and has been applied in 16 plants, mainly for smelter gas and tail gas from sulfuric acid plants. It has also been tested for flue gas from coal at the Tennessee Valley Authority's Shawnee Power Station (9). In 1983 and 1984, three plants went into operation, including a plant in North Korea which has a treatment capacity of 50,000 Nm³/h for flue gas from a sintering machine.

A recent improvement in the process is the use of a hydrocyclone to replace the thickener, thus reducing investment cost and the amount of space required. Operational parameters for one of the new plants are shown in Table 3-16.



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Figure 3-12. FGD plant using the converter slag process at Sakai, Nippon Steel Co. (two units, each 550,000 Nm³/h).

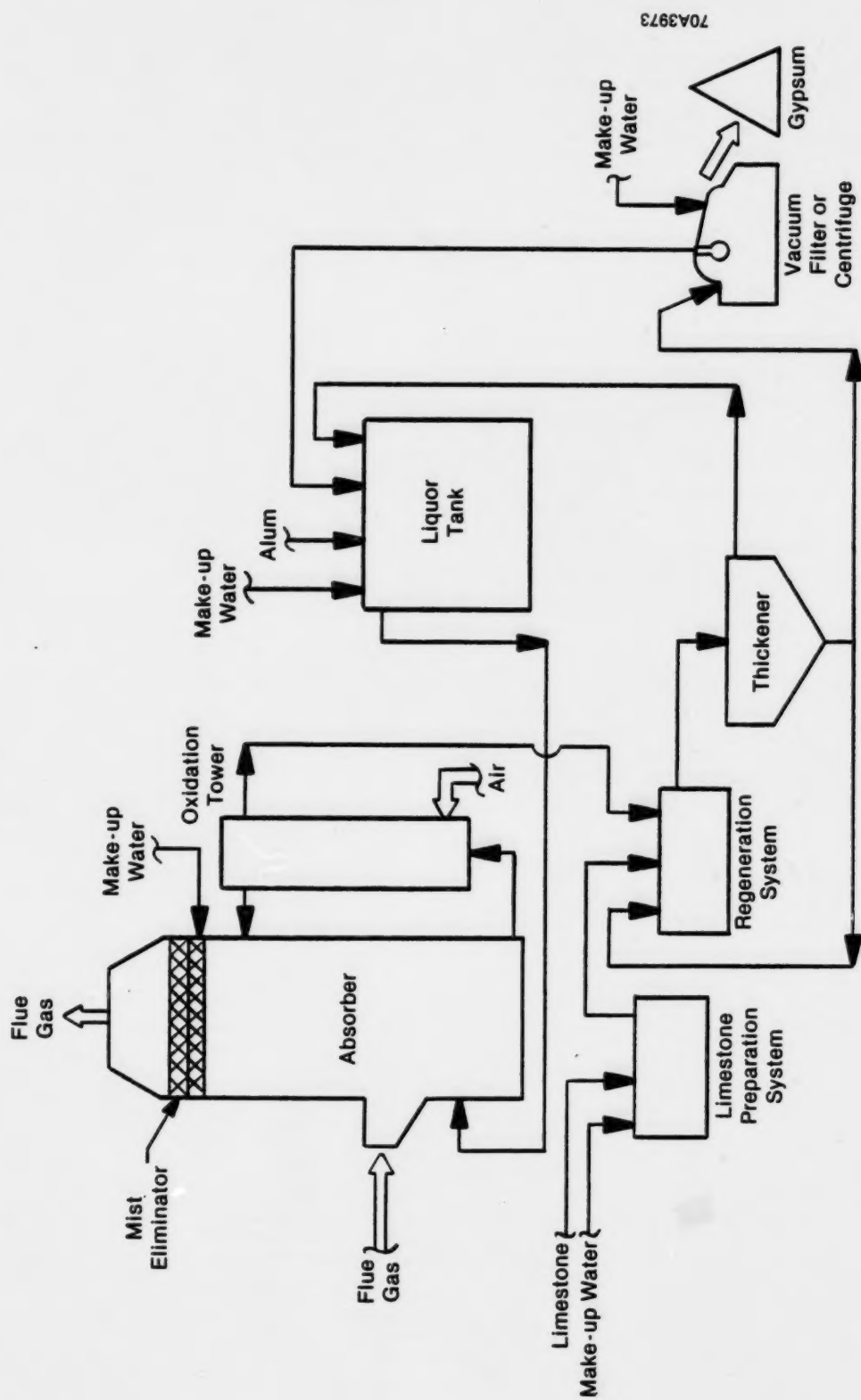


Figure 3-13. Flowsheet of the Dow process.

3.8.5 Mitsubishi Petrochemical's Yokkaichi Plant (Chiyoda CT-121 Process)

The plant is the first commercial application of Chiyoda's CT-121 process using the Jet Bubbling Reactor that was described in Section 3.5. The plant has a flue gas treatment capacity of 250,000 Nm³/h from an industrial boiler that burns a high sulfur oil to produce 280 t/h steam (85 MW equivalent). A flowsheet for the plant would be similar to that of the Toyama plant (refer to Figure 3-4), except that the Yokkaichi plant has no gas-gas heater in the flue gas reheating system. The flue gas is introduced into a jet bubbling reactor after being treated by a small venturi-type prescrubber. The reactor has 800 gas spargers made of a plastic material and contains a slurry at pH 3.5.

A typical example of the operational parameters at full load is shown in Table 3-16. A high level of SO₂ removal efficiency, 98%, was attained with a low power consumption of 1.3 percent. Example operational data at a reduced load are as follows: the flue gas volume of 189,000 Nm³/h contained 1,890 ppm SO₂, which was reduced to 40 ppm while the dust was reduced from 100 mg/Nm³ to 30 mg/Nm³. The pressure drop was about 300 mm H₂O across the reactor and 15 mm H₂O across the mist eliminator. Power consumption was 1,030 kW, accounting for 1.6 percent of the power generated. Water consumption was 20 t/h, yielding 0.8 t/h wastewater.

Inspection after one year of operation showed that 2 or 3 spargers (out of 800) had been broken due to problems in fabrication but the breakage caused no operational difficulties. No other problem has been experienced. The operation is easy and requires only part of one person's time per shift, excluding gypsum handling. Gypsum grows into a large crystal (around 100 microns), allowing the use of a continuous centrifuge to reduce moisture content 11-12%. The gypsum has been sold for cement and wallboard for 3,000 yen/t, including about 1,000 yen/t for transportation. The moisture content of the gypsum may be 6-7% when a batch centrifuge is used.

The FGD plant cost 1.5 billion yen (18,000 yen/kW). Maintenance costs have been less than 5 million yen/year, including inspection costs. The operating cost is about 80 yen/ton of steam generated by the boiler, which is about one-half that of the Chiyoda 101 process using dilute sulfuric acid scrubbing.

SECTION 4

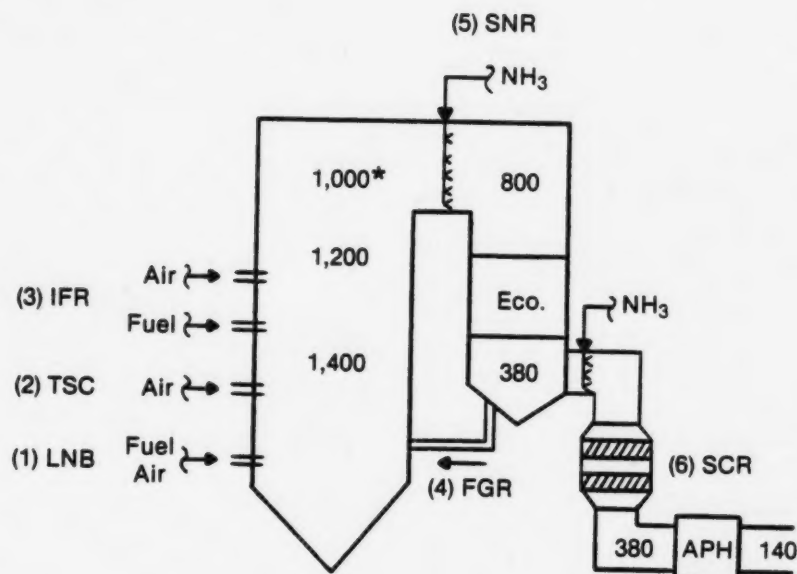
NO_x ABATEMENT BY COMBUSTION MODIFICATION (CM)

4.1 NO_x ABATEMENT FOR BOILERS

NO_x abatement methods for boilers are illustrated in Figure 4-1. Among the methods, combustion modification (CM) including a low-NO_x burner (item 1 of the figure), two-stage combustion, item 2, and flue gas recirculation, item 4, have been most widely used for NO_x reductions of 20-80%. NO_x concentrations in flue gases from utility boilers without CM reach 600-900 ppm for coal-firing, 300-400 ppm for oil-firing, and 200-300 ppm for gas-firing. Through the use of CM, NO_x concentrations have been lowered to 150-300 ppm for coal (at 6% O₂ basis, for dry bottom), 60-110 ppm for oil (at 3% O₂ basis), and 40-80 ppm for gas (at 4% O₂ basis).

For further NO_x abatement, in-furnace NO_x reduction (IFR, or three-stage combustion, or reburning, item 3 of Figure 4-1), has been developed in Japan and the U.S. In this technology, a small portion of the fuel for the boiler is injected above the flame to reduce NO_x by 40-50% of the NO_x (see also Section 6.3).

For NO_x removal from flue gas, selective noncatalytic reduction (SNR, item 5 of Figure 4-1) and selective catalytic reduction (SCR, item 6 of the figure) have been developed. Both use ammonia for NO_x reduction. In the SNR process, ammonia is injected at 800-1,000°C to remove 30-50% of the NO_x. This will be discussed in more detail in Section 6.2. SCR uses ammonia and a



* Numbers Indicate Gas Temperature, °C

- | | |
|---|---|
| (1) Low- NO_x burner | (5) Selective noncatalytic reduction (Thermal DeNO_x) |
| (2) Two-stage combustion | (6) Selective catalytic reduction |
| (3) In-furnace reduction | Eco.: Economizer |
| (4) Flue gas recirculation (Gas mixing) | APH: Air preheater |

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Figure 4-1. NO_x abatement methods for utility boilers.

catalyst at temperatures of 300-400°C to remove 60-90% of the NO_x (see Section 5). SCR often is used in combination with combustion modification.

4.2 FUNCTIONS OF CM TECHNIQUES

NO_x formed by combustion consists of thermal NO_x from the oxidation of nitrogen in the air at high temperatures and fuel NO_x , which is derived from nitrogen compounds in the fuel. Generally, the former is predominant for oil and gas combustion and the latter is predominant for coal combustion. Major techniques of CM together with their functions, effects, and problems are summarized in Table 4-1. Thermal NO_x can be reduced by various CM techniques, such as reducing the O_2 concentrations in the combustion regions and lowering the temperature. Fuel NO_x can be reduced by lowering the O_2 concentration in the initial combustion zone through staged combustion, or fuel switching.

Many combinations of the above techniques have also been used. Although operating conditions can be changed quite easily in existing installations, this does not usually reduce NO_x emissions significantly and often causes operating problems instead. Modifications in burner and combustion system design are promising control techniques, but the application of these techniques to existing installations is not as easy as modifying the operating conditions. Fewer problems are encountered when design modifications are incorporated in new plants. Relatively large reconstruction costs are required when system design modifications are applied to existing installations.

In Japan, NO_x emission controls have been applied to over 100,000 sources including boilers, furnaces and kilns, heaters, and incinerators, representing about 95% of the total emissions from all stationary sources. Virtually all of the sources use combustion modification for NO_x abatement. Smaller sources reduce NO_x by 20-50% mainly through the use of the low- NO_x burner method or by fuel switching, while large sources use the combination of two-stage combustion, flue gas recirculation, and low- NO_x burners to reduce NO_x 50-80%.

TABLE 4-1. CLASSIFICATION OF CH TECHNIQUES AND THEIR FUNCTIONS

Type	CH Technique	Major Function	Effect on		Problems	Ease of Application to Existing Furnaces
			Thermal NO _x	Fuel NO _x		
Operating condition modifications	Low excess air combustion	Reduces O ₂ in flue gas	⊙	0	Tends to cause incomplete combustion	Easy
	Reduction of heat load in furnace	Lowers combustion temperature	⊙	X	Reduces capacity	Easy
	Lowering of air preheating temp.	Lowers combustion temperature	⊙	X	Energy loss	Possible
Combustion system modifications	Two-stage combustion	Initiates combustion in O ₂ deficient condition	⊙	⊙	Incomplete combustion Extension of flame	Possible
	Flue gas recirculation	Lowers combustion temperature Lowers O ₂ concentration	⊙	0	Increases cost Expansion of flame	Possible
	Low-NO _x burner	Various	Various	Various	Various	Possible
Others	Water or steam injection	Lowers temperature Lowers O ₂ concentration	⊙	X	Reduces thermal efficiency	Possible
	Fuel switching	Reduces N in fuel	0	⊙	Increases cost	Difficult in some cases

⊙ Effective.

0 Some effect.

X Little or no effect.

Table 4-2 shows NO_x abatement for boilers using various methods of combustion modification. NO_x concentrations in utility boiler flue gases have been lowered by combustion modification to 150-350 ppm for coal-firing, 60-130 ppm for oil-firing, and 40-80 ppm for gas-firing to meet the national emission standards and most local regulations. Selective catalytic reduction (SCR) of NO_x is sometimes applied in addition to combustion modification when the local regulations are so stringent that they cannot be met by combustion modification alone.

TABLE 4-2. COMBUSTION MODIFICATION AND NO_x ABATEMENT

	Fuel N Content (%)	Coal ^a	Oil	Gas
		0.7-3.0	0.1-0.5	0
Combustion Modification	No control	500-800	250-400	200-300
	Flue gas recirculation (FGR)	400-600	180-300	150-200
	Two-stage combustion (TSC)	300-500	180-300	150-200
	FGR + TSC	250-400	120-250	100-150
	FGR + TSC + low-NO _x burner	150-300	60-150	40- 80

^aPulverized coal, dry bottom.

4.3 COMBUSTION MODIFICATION FOR OIL- AND GAS-FIRING

4.3.1 Overview

Almost all of the NO_x from gas-firing and 60-80% of the NO_x from oil-firing are derived from thermal NO_x, which can be reduced relatively easily by such principles of combustion modification as lowering the combustion temperature, shortening the retention time at high temperature, and reducing the O₂ concentration. To attain those conditions, flue gas recirculation and two-stage combustion are applied and various types of low-NO_x burners have been developed.

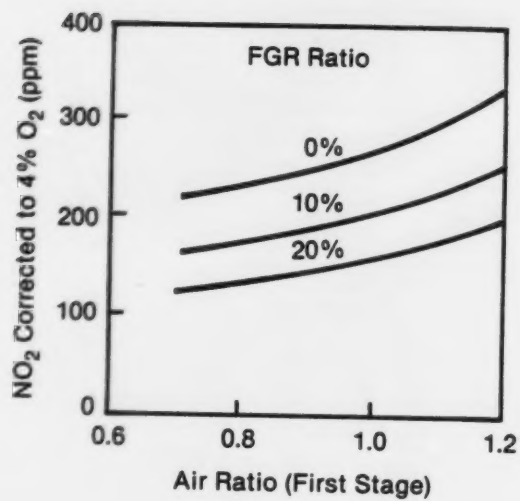
Figure 4-2 shows a typical example of the effect of flue gas recirculation (FGR), two-stage combustion (TSC), and their combined use for an oil-fired boiler. Without combustion modification, NO_x concentrations may reach 350 ppm (0% FGR, first stage air ratio 1.2). With 20% FGR, NO_x was reduced to 200 ppm. By applying TSC without FGR and using the air ratio of 0.8 in the first stage, NO_x was reduced to 230 ppm. With TSC and 20% FGR, NO_x was reduced to 130 ppm. NO_x might be further reduced by intensifying FGR and TSC, for example, by applying 30% FGR and using an 0.7 air ratio at the first stage. Such intensification, however, often causes problems as was shown in Table 4-1.

4.3.2 Low- NO_x Burner for Oil- and Gas-firing

More than 20 manufacturers of low NO_x burners in Japan produce more than 50 types of low- NO_x burners for commercial use. Most of the burners are for oil- or gas-firing. These burners may be classified into the following categories, according to the principles of NO_x abatement employed (10).

- (1) High-mixing
- (2) Divided-flame
- (3) Self-recirculation
- (4) Two-stage combustion
- (5) Off-stoichiometric combustion
- (6) Combination
- (7) Water-injection

The high-mixing type burner reduces the formation of NO_x by rapid mixing of the fuel and air and the use of rapid combustion to shorten the residence time in a high temperature zone. The divided flame burner divides the flame into several small flames by means of nozzles with grooved tips so that the heat is released rapidly and the flame temperature lowered. In the self-recirculating burner, a small portion of the combustion gas is recirculated by the jet force to lower both the O_2 concentration in the combustion zone and



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Figure 4-2. Effect of flue gas recirculation (FGR) and two-stage combustion. (utility boiler, C-grade heavy oil, total air ratio: 1.2)

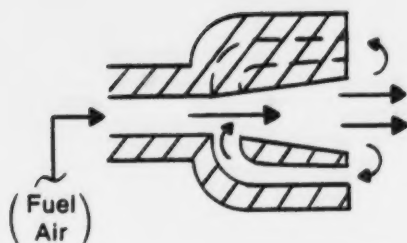
also the flame temperature (Figure 4-3, A). The two-stage combustion burner gives a fuel-rich flame which is mixed with air in the second combustion stage (Figure 4-3, B). The off-stoichiometric burner gives a fuel-rich flame and air-rich flames which mix for complete combustion and low NO_x formation (Figure 4-3, C). The water injection burner injects water to lower the combustion temperature. In addition, there are many types of burners that utilize self recirculation, two-stage combustion, and water injection. These burners usually cost 30-60% more than conventional burners and reduce 20-40% of the NO_x . Higher grade burners that are more costly but able to reduce more than 50% of the NO_x with a combination of the above techniques have also been developed (Figure 4-3, D).

4.3.3 Example Utility Application

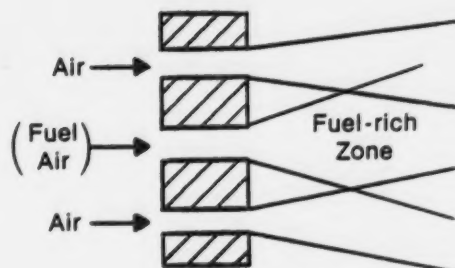
Table 4-3 shows Tokyo Electric Power Co.'s application of combustion modification to oil- and gas-fired boilers. A combination of flue gas recirculation, two-stage combustion, and a low- NO_x burner is popular and lowers the NO_x concentrations to about 110 ppm and 80 ppm for oil- and gas-fired boilers, respectively.

TABLE 4-3. TOKYO ELECTRIC'S OIL- AND GAS-FIRED BOILERS APPLYING COMBUSTION MODIFICATION (April 1981)

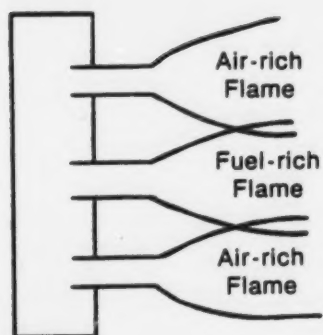
Methods of Combustion Modification	Number of Boilers	Total Generating Capacity (MW)
Flue gas recirculation (FGR) only	15	1,857
FGR + Two-stage combustion (TSC)	12	5,289
FGR + Low- NO_x burner (LNB)	9	2,710
FGR + TSC + LNB	<u>32</u>	<u>13,605</u>
Total	68	23,461



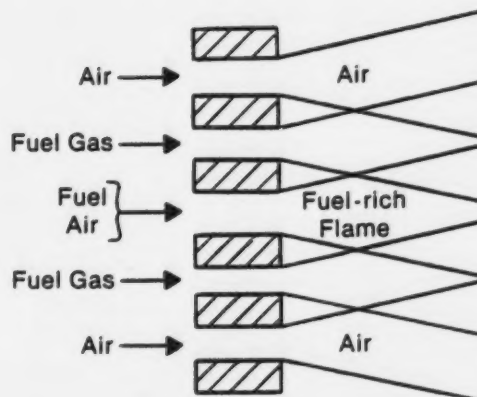
(A)
Self-recirculation
Type



(B)
Two-stage
Combustion Type



(C)
Off-stoichiometric
Combustion Type



(D)
Combination Type
(FGR + TSC)

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Figure 4-3. Four low- NO_x burner types.

Table 4-4 shows the regulations and level of NO_x abatement for oil- and gas-fired boilers at the Chita Station of Chubu Electric and the Kudamatsu Station of Chugoku Electric. All of the boilers use combustion modification to meet the emission standards of the central government. Most of the boilers apply denitrification, mainly selective catalytic reduction, to meet local regulations.

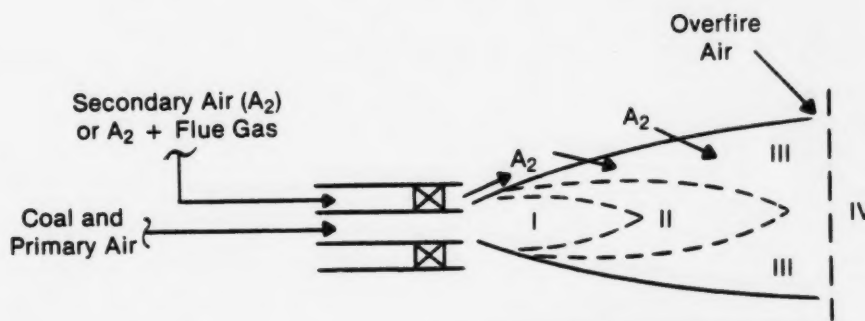
4.4 COMBUSTION MODIFICATION FOR COAL-FIRED BOILERS (11)

4.4.1 Principles of NO_x Abatement for Pulverized Coal Burning

For pulverized coal combustion, the reduction of fuel NO_x is of particular importance because coal contains much larger amounts of nitrogen (1-3%) than does oil (0.1-0.5%) and produces a large amount of fuel NO_x , accounting for 70-80% of the total NO_x . The fuel NO_x from coal consists both of volatile NO_x (derived from volatile nitrogen compounds in the coal) and char NO_x (derived from residual nitrogen compounds in the char). Usually the former is more predominant than the latter. The volatile NO_x can be abated efficiently by combustion control using a low- NO_x burner (LNB). Char NO_x is not controlled as easily, since it is less sensitive to injection design modifications in the early stages of combustion. Staged combustion (overfire air) is highly effective for NO_x abatement for pulverized coal because it reduces both fuel and thermal NO_x . However, flue gas recirculation (gas mixing) is not as effective because it mainly reduces thermal NO_x , which is not as much a problem for coal-burning facilities.

A low- NO_x combustion model of pulverized coal by a LNB and staged combustion is shown in Figure 4-4. Usually 70-90% of total air for complete combustion is injected through the burner, with the rest through the overfire air port. In the primary combustion zone (I), volatile matter begins to burn, producing much NO_x due to the combustion of volatile nitrogen compounds.

In the secondary zone (II), the volatile matter continues to burn, the char begins to burn, and O_2 concentrations decrease, producing a reducing



Zone	Burning Matter	NO_x Concentration
I	Most of volatile matter	High
II	Remaining volatile matter Part of Char	Lowered (reducing atmosphere)
III	Char	Minimized (reducing atmosphere)
IV	Remaining char Reducing gas (CO , etc.)	Increased (oxidizing atmosphere)

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Figure 4-4. Model of low- NO_x coal burning.

TABLE 4-4. NO_x CONTROL FOR OIL- AND GAS-FIRED UTILITY BOILERS

	Chubu Electric (Chita Station)						Chugoku Electric (Kudamatsu Station)		
	1	2	3	4	5	6	1	2	3
Boiler No.	375	375	500	700	700	700	156	375	700
Capacity, MW	MHI ^a	MHI	MHI	MHI	BHK ^b	BHK	IHI ^c	IHI	IHI
Boiler mfr.	1965	1966	1967	1974	1978	1978	1964	1973	1979
Operation start-up	Oil	Oil	Oil	Oil	Gas	Gas	Oil	Oil	Oil
Fuel	120-140	120-140	90-100	95-100	40-50	40-50	140-150	140-150	110-130
Boiler outlet NO _x , ppm ^d	180	180	180	180	100	100	180	180	130
NO _x limitation, ppm	60	100	105	20	11	11	160	32	26
By Central Government									
By Local Government									
Denitrification plant									
Process	SCR ^e	SNR ^f	-	SCR	SCR	SCR	-	SCR	SCR
Constructor	MHI	MHI	-	MHI	Hitachi ^g	Hitachi	-	IHI	IHI
Completion	1983	1977	-	1980	1978	1978	-	1979	1979
NO _x removal, %	60	30-40	-	80-85	80	80	-	81-88	81-83
Outlet NO _x , ppm	60	80-100	90-100	15-20	8-10	8-10	140-150	24-28	20-24
Outlet NH ₃ , ppm	Below 3	10-15	Below 5	Below 5	Below 5	Below 5	-	Below 5	Below 5

^aMitsubishi Heavy Industries

^bBabcock Hitachi K. K.

^cIshikawajima-Harima Heavy Industries

^dBy combustion modification

^eSelective catalytic reduction

^fSelective Noncatalytic reduction

^gHitachi Ltd.

atmosphere that reduces NO_x to N_2 . The reducing atmosphere also exists in the tertiary zone (III), where the NO_x concentrations reach a minimum just before the overfire air port for staged combustion. The CO concentration remains high. When the staged air (IV) is injected, the remaining char, CO, and other reducing gases are burned, resulting in a slight increase in NO_x concentration.

To minimize the NO_x level, the gas retention time in the reducing atmosphere should be as long as possible and the mixing of the secondary air with the flame formed by coal and primary air should be delayed. The resulting long flame with extended reducing zone, however, may promote slag formation and boiler tube wastage. Also, the flame may be unstable and unburned carbon may increase. Countermeasures for such problems should be taken, otherwise, the NO_x reduction should be limited to where it does not cause an obvious problem.

Many LNBs have been developed to achieve a low NO_x level for various types of coal. Air ratios, coal injection velocity, flame swirling velocity, etc., are adjusted according to the nature of coal and the type of the boiler. Some LNBs use recirculated flue gas which is mixed with the secondary air or injected between the secondary air and the flame; this may increase NO_x abatement efficiency, but it increases the cost because of the required flue gas recirculation facility.

Examples of NO_x abatement by low- NO_x burners at coal-fired utility boilers are shown in Table 4-5.

LNBs may cost roughly 50% more than conventional burners. Yet, NO_x abatement by a LNB and CM is much less costly than by flue gas treatment, primarily because of relatively low operating costs. Even when selective catalytic reduction (SCR) of NO_x is applied for extensive NO_x abatement, it is generally more economical to use a LNB and CM with SCR than to use SCR alone.

TABLE 4-5. EXAMPLES OF COMBUSTION MODIFICATION FOR COAL-FIRED UTILITY BOILERS

Station (Company)	Boiler (MW)	Coal Source	Burner Type	NO _x (ppm)
Isogo (EPDC)	265 ^a	Domestic ^b	DF-CN ^c	150-200
Tomato-Atsuma (Hokkaido)	350 ^d	Domestic ^b	DRB ^e	200
Matsushima (EPDC)	500 ^d	Imported	SGR ^f	200-280
Sunagawa (Hokkaido)	125 ^d	Domestic ^b	VDG	160-170

^a Staged combustion used

^b Low N, high volatile

^c Dual flow convergent fuel nozzle burner

^d Staged combustion and flue gas recirculation used

^e Dual-air-register burner

^f Separate-gas recirculation burner

^g Vortex diffuse burner

Possible problems with a LNB and CM for coal-fired boilers include increased unburned carbon, slag formation, and tube wastage. Countermeasures should be taken to solve the problems. For a wet-bottom boiler operated at higher temperature to fuse the ash, extensive NO_x abatement by a LNB and CM is not easily achieved. The following subsections describe NO_x abatement by LNBs and CM for dry-bottom utility boilers.

4.4.2 EPDC's Tests on Low-NO_x Coal Combustion (11, 12)

The Electric Power Development Company (EPDC) has conducted extensive tests of combustion modification for NO_x abatement, with major boiler manufacturers including Mitsubishi Heavy Industries (MHI), Babcock Hitachi, Ltd. (BHK), Ishikawajima-Harima Heavy Industries (IHI), and Kawasaki Heavy Industries (KHI). Some of the test results shown in Figure 4-5 indicate that, by using a combination of low-NO_x burners with staged combustion and flue gas

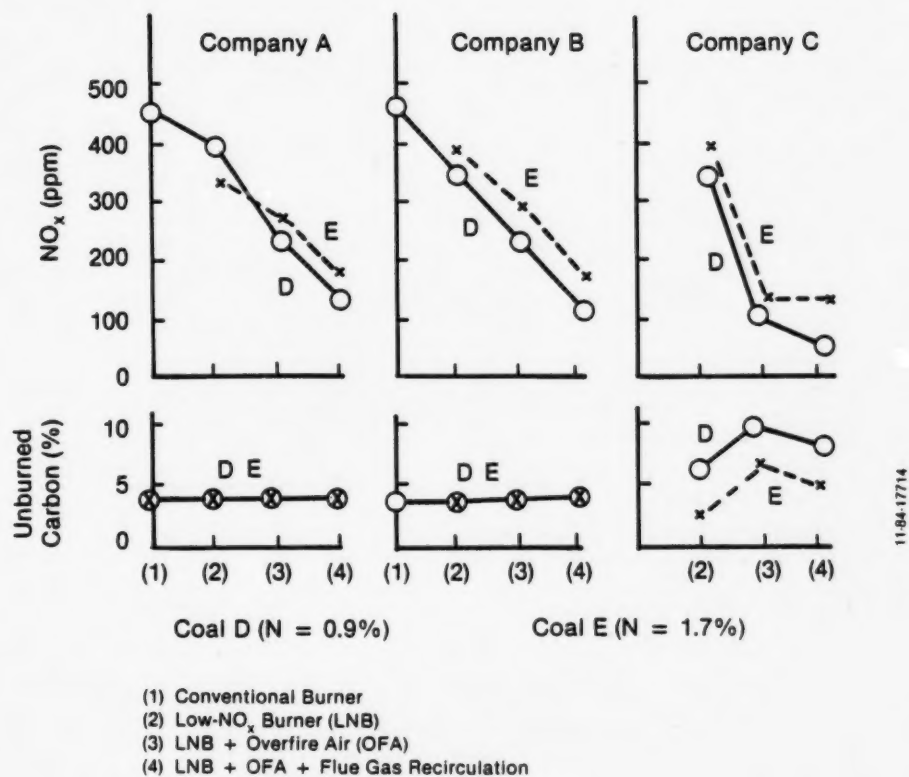


Figure 4-5. Examples of combustion modification tests conducted by EPDC and boiler manufacturers.

recirculation, NO_x can be reduced by 60-80% (to 100-150 ppm with low-nitrogen coal and to 150-200 ppm with nitrogen-rich coal). A test conducted jointly with Company C indicated that flue gas recirculation was not effective for NO_x abatement with Coal E, and that unburned carbon increased considerably with the application of staged combustion.

4.4.3 NO_x Abatement by DF-CN Burner at EPDC's Isogo Station (12, 13, 14)

IHI and Foster Wheeler Energy Corporation of the U.S. have developed a low- NO_x burner called the IHI-FW DF-CN (Dual Flow Convergent Fuel Nozzle) burner. It is based on the following principles for NO_x reduction:

- 1) Minimum primary/secondary mixing prior to completion of the combustion of the volatile fraction;
- 2) Ignition stability at the injector; and
- 3) Dilution of the secondary air with self-recirculating combustion products.

The burner has been used at EPDC's Isogo Station for two 265 MW coal-fired boilers (IHI-FW single drum type with natural circulation) which burn domestic low-nitrogen coal (about 1.0 percent N), rich in volatile matter (about 40%). Since the station is located in Yokohama City with a population of about 5 million, increasingly stringent NO_x control has been required by the city.

The boilers were completed in 1967 and 1969, and were modified in 1973 with an overfire air port system for NO_x abatement (Table 4-6). As a second step to meet the increasingly stringent NO_x regulation, an IHI-FW DF-CN burner and a boundary air system (Figure 4-6) were installed in 1977. The boundary air system prevents the melting of the ash. As a third step, eight additional overfire air ports were installed in 1979 near the side walls and near a division wall, based on tests conducted by IHI. In this way, NO_x was reduced

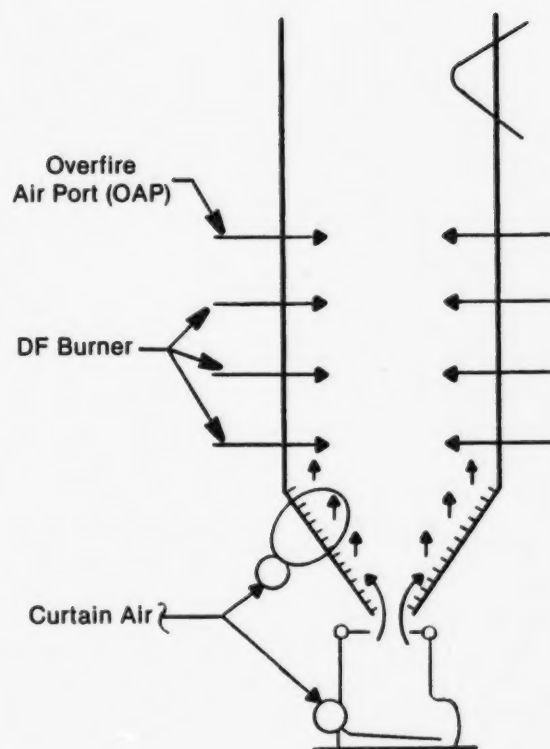


Figure 4-6. IHI's Boundary Air System.

to 170-190 ppm by opening the overfire air damper about 40 percent (overfire air nearly 20%). In April 1980, further NO_x reduction to 159 ppm was required by the city. This NO_x reduction has been attained by increasing the damper opening ratio. Tests on steam injection through oil-burner guns indicated a little NO_x reduction at a small damper opening ratio (Figure 4-7).

TABLE 4-6. MODIFICATIONS MADE FOR INCREASED NO_x CONTROL AT THE ISOGO POWER UNIT

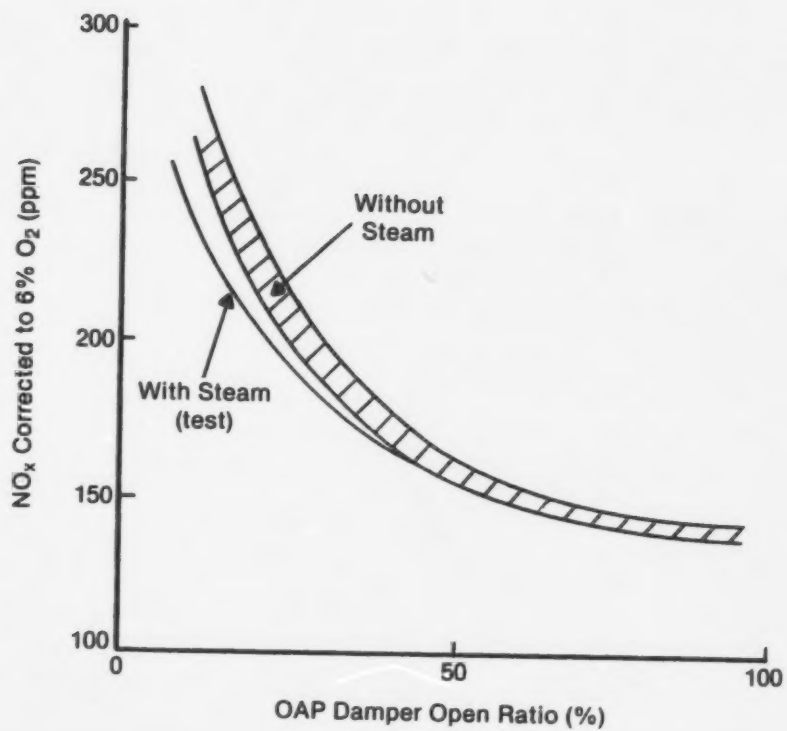
Month, Year	NO _x Emission Level by Regulation (ppm) ^a	Achieved NO _x Level (ppm)	Modifications
Before May 1973	600	570	None
June 1973	600	380 - 510	Two-stage combustion
April 1977	480	240 - 250	Low-NO _x burner, curtain air
April 1979	400	170 - 190	More rigorous two-stage combustion

^aBy Central Government

4.4.4 DRB and PG Dual Burner at EPDC's Takehara Station (12, 15, 16)

The Takehara station has two Babcock and Wilcox coal-fired boilers--No. 1 (250 MW) and No. 3 (700 MW) -- constructed by BHK. A dual-air-register burner (DRB) developed by Babcock & Wilcox, U.S.A., is used for boiler No. 1, and a primary gas (PG) dual burner is used for boiler No. 3. The PG dual burner is based on the dual-air-register burner with improvements by BHK. The major improvement is that the PG dual burner has a primary gas port for injecting recirculated flue gas between the secondary air and the flame formed by coal and the primary air.

Figure 4-8 shows the relationship between the heat release rate at the burner zone and the resulting NO_x concentrations with three types of



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Figure 4-7. Damper opening ratio and NO_x concentration (Isogo).

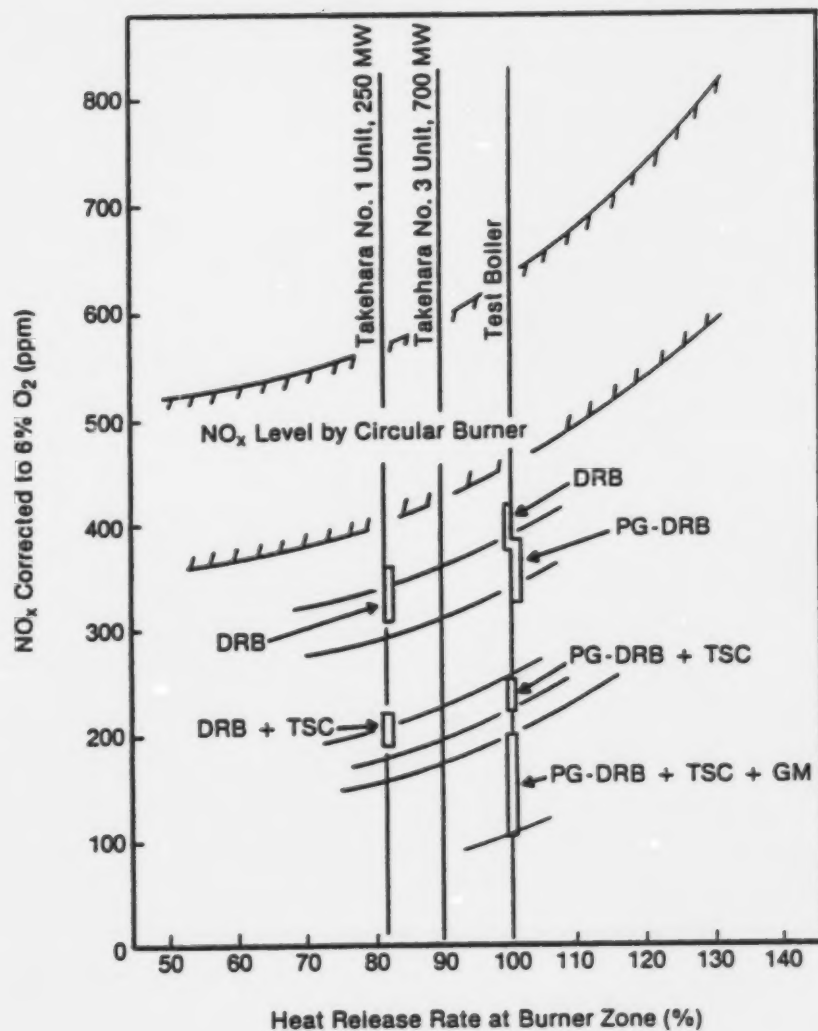


Figure 4-8. Heat release rate at burner zone and NO_x concentration.

burners--conventional circular-air register, dual-air register, and PG dual burner with or without two-stage combustion and gas mixing. Boiler No. 1 uses a dual-air register burner and produces about 200 ppm NO_x using two-stage combustion, and about 300-360 ppm NO_x without two-stage combustion. A test furnace with a higher test release rate produced about 400 ppm NO_x with the dual-air register burner and around 350 ppm NO_x with the PG dual burner when staged combustion was not applied; it produced 100-200 ppm NO_x with the PG dual burner with two-stage combustion and gas mixing.

Boiler No. 3, with a heat release rate between that of boiler No. 1 and the test furnace, may be able to produce a NO_x concentration of about 150 ppm. Actually, the NO_x level from boiler No. 3 has been kept at 250-300 ppm because the flue gas is further treated by selective catalytic reduction to reduce NO_x to about 60 ppm.

4.4.5 SGR Burner and PM Burner (12, 17, 18)

MHI has developed a separate-gas recirculation (SGR) burner which is used at EPDC's Matsushima Station for two 500 MW boilers. The burner is based on two principles that MHI has found to be important for NO_x abatement:

- (1) The temperature in the volatile matter combustion zone should be kept as high as possible, and combustibles should stay in this zone as long as possible.
- (2) The temperature of the char combustion zone should be appropriately low, and combustion should be completed in this zone.

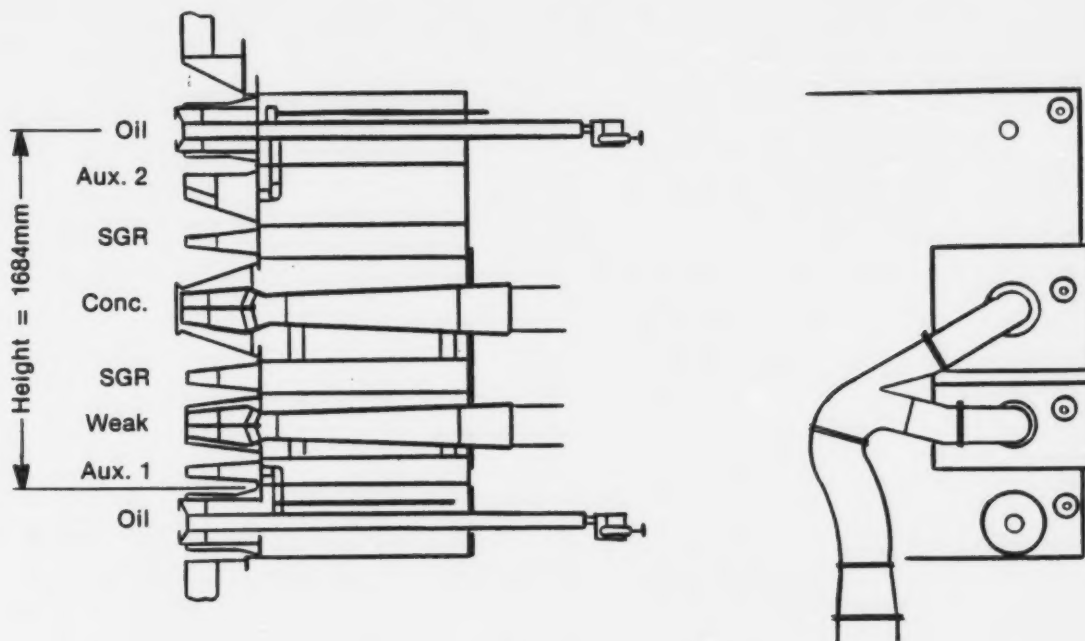
The burner injects recirculated flue gas between secondary air and the flame formed by the coal and primary air. A flame holder at the outlet of the coal nozzle maintains stable ignition and expedites the combustion of volatile matter. NO_x concentrations in flue gases from the tangential-firing 500 MW boilers have been maintained at 200-280 ppm with imported coals.

For further NO_x abatement, MHI has developed the Pollution Minimum (PM) burner which gives a fuel-rich flame and a fuel-lean flame with recirculated flue gas injected in between to conduct off-stoichiometric combustion efficiently (Figure 4-9). Figure 4-10 shows the relation between NO_x concentrations and the primary air/coal ratio in kg/kg. A ratio of about 3-4 corresponds to the theoretical air for volatile matter in coal. NO_x concentrations are lowered with the smaller ratio because of the insufficient O_2 . On the other hand, the ratio 7-8 corresponds to theoretical air for coal. With the ratio of between 3-4 and 7-8, the fixed carbon in coal burns with insufficient O_2 to form NO_x .

A conventional burner (C_0) may produce a primary air/coal ratio of 2-3 as shown in Figure 4-10. If the primary air/coal supply is divided into two streams with different ratios-- C_1 for a coal-rich concentrated nozzle and C_2 for coal-lean weak nozzle--for separate burning, the average NO_x value should be $(\text{NO}_x)\text{PM}$ as shown in Figure 4-10. This is the weighted mean of $(\text{NO}_x)\text{C}_1$ and $(\text{NO}_x)\text{C}_2$ and is substantially lower than $(\text{NO}_x)\text{C}_0$. The combined use of the concentrated weak flames may result in low- NO_x combustion.

Figure 4-11 compares the NO_x concentrations for a Combustion Engineering (CE) conventional burner, an SGR burner, and a PM burner when used for tangential-firing commercial boilers and the test furnace. Since NO_x concentrations for test boilers are very similar to those for commercial boilers, it is expected that with use of a commercial PM burner, NO_x concentrations may be reduced to 60-80 ppm with Japanese coals and to 100-150 ppm with imported coals.

Since late 1982, the PM burner has been applied to a 100 MW industrial boiler of Daishowa Paper Co. that uses Australian coal. By increasing overfire air from 5 to 17%, the NO_x concentration decreased from 210 to 150 ppm with little increase in unburned carbon--from about 4 to 4.5% (19).



(Aux. 1, Aux. 2: Secondary Air
 SGR: Recirculated Flue Gas
 Conc: Fuel-rich Flame
 Weak: Fuel-lean Flame)

Figure 4-9. MHI PM burner.

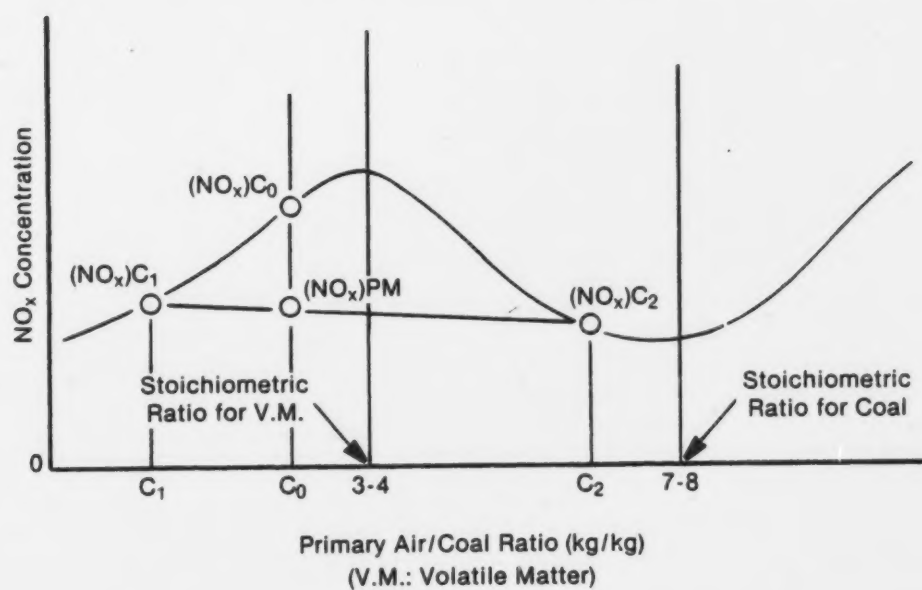


Figure 4-10. Concept of pulverized coal-fired low- NO_x PM burner.

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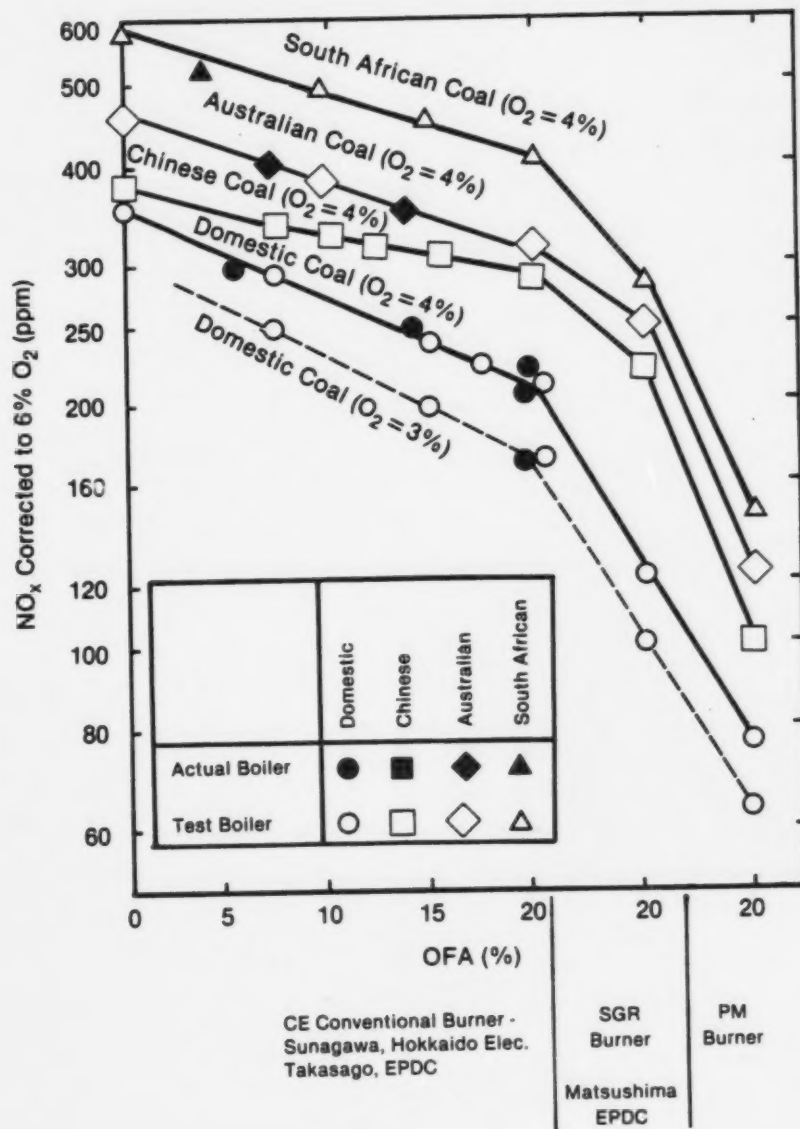


Figure 4-11. Comparison of NO_x emissions: laboratory test results vs. field data.

4.4.6 VD Burner (20)

KHI has developed a low- NO_x burner called the vortex diffuse (VD) burner which is used at Hokkaido Electric's Sunagawa Station (Table 4-5). The burner is designed to reduce NO_x efficiently when overfire air is added, and to prevent any increase in unburned carbon. The swirl vane in the primary throat has a relatively large angle to ensure flame holding and to widely distribute pulverized coal in the furnace to increase combustion efficiency. Secondary and tertiary air is introduced to provide slow mixing with the coal and the primary combustion products by the deviation effect of the flame holder. Recirculated flue gas can be mixed with the secondary and tertiary air for further NO_x abatement. By using the VD burner with 25% overfire air, NO_x has been reduced to 160-170 ppm with domestic coal which is low in nitrogen and rich in volatile matter. This coal is similar to that used at EPDC's Isogo Station.

4.5 COMBUSTION MODIFICATION FOR VARIOUS FURNACES

4.5.1 Cement Kilns

Rotary kilns for cement production produce a flue gas rich in NO_x -- 500-1,000 ppm at 10% O_2 . This is due to the long retention time of the gas at temperatures of 1,000-1,500°C and to the presence of a large excess of air. The long retention time is necessary for the calcination of raw materials (limestone, clay, etc.) and for the reaction between the calcination products.

A new suspension preheater kiln (NSP) with a furnace (DD furnace) between the rotary kiln and the suspension preheater has been developed in Japan. Figure 4-12 shows this process at Nihon Cement Co.'s Kamiiso plant which has a capacity of producing 3,800 t/day of cement clinker. Process specifications are shown in Table 4-7. About half of the total fuel is injected into the DD furnace through the main burner to calcine the raw materials. This reduces the heat load in the rotary kiln and thus NO_x is reduced to about 200 ppm for oil-firing and 400 ppm for coal-firing to meet the regulations, while cement

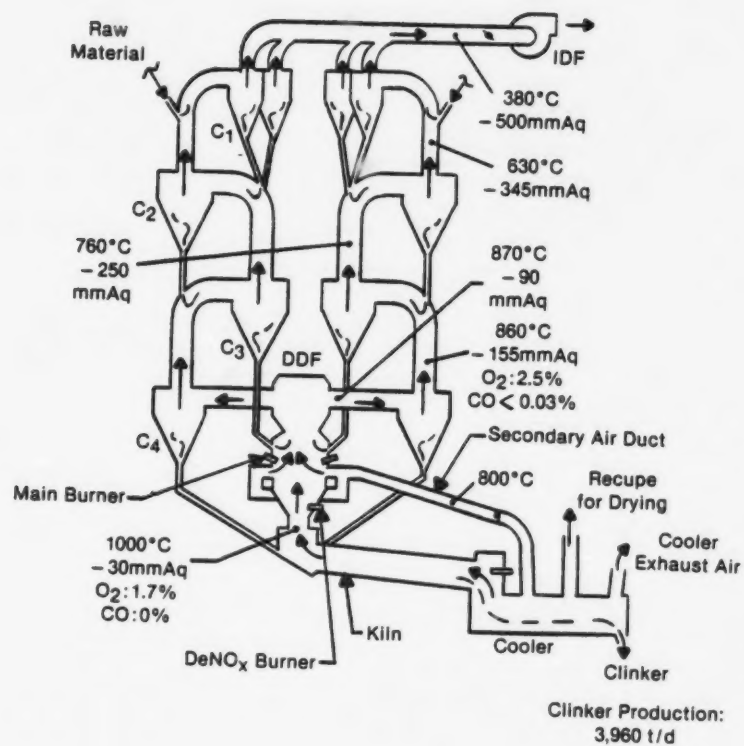


Figure 4-12. DD process flow and operating data.

production capacity is increased. If necessary, NO_x can be further reduced by the use of the DeNO_x burner, as will be described in Section 6.3.

TABLE 4-7. PRINCIPAL SPECIFICATIONS OF THE DD PROCESS

Preheater	Type: Suspension Preheater with DD Furnace Capacity: 3,600 t/d Structure: 20(l) x 15(w) x 64(h)m C_1^a : 3.8 m x 4 C_2 : 5.9 m x 2 C_3 : 6.4 m x 2 C_4 : 6.4 m x 2 DDF: 6.0 m x 1
Kiln	Capacity: 3,600 t/d 4.55 x 76.7 ¹ m x 3.5 %
Cooler	Type: Horizontal Type Fuller 1,230/1,460 Effective Area: 110 m ² Capacity: 3,800 t/d

^aLetters refer to those found on Figure 4-12

4.5.2 Glass Melting Furnaces

Glass melting furnaces usually burn oil or gas and emit high levels of NO_x (about 500 ppm) due to 1) a high furnace temperature (about 1,600°C), 2) a relatively long gas retention time which is needed to ensure the melting and clarification of the glass, and 3) the high O_2 concentration of the gas. When sodium nitrate is added as a feed to promote clarification, the NO_x concentration in the flue gas commonly exceeds 2,000 ppm. The use of nitrates has

been prohibited in Japan to reduce NO_x emissions. In furnaces not using nitrate, virtually all of the NO_x is due to thermal NO_x , and can be lowered in much the same manner as it is in other oil- and gas-fired furnaces. In addition, prevention of air leakage is important for glass furnaces.

An oil-pressure atomizer that produces a plane flame shape has been developed by the Asahi Glass Co. The plane flame shape is formed by a slit in the atomizer tip. With the combination of this atomizer and an adequately shaped port, a 40% NO_x reduction can be achieved.

4.5.3 Refuse Incinerators

Flue gas from refuse incinerators usually contains 150-200 ppm NO_x at a 12% O_2 basis. Most of the NO_x (70-80%) is derived from nitrogen compounds in the refuse. Therefore, two-stage combustion is effective to reduce NO_x by 30-40%, similar to NO_x abatement for coal-fired boilers. Problems with the two-stage combustion design and modifications to remedy them are shown in Table 4-8.

TABLE 4-8. PROBLEMS AND SOLUTIONS WITH TWO-STAGE COMBUSTION FOR REFUSE INCINERATORS

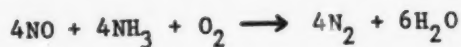
Problems	Modifications for Solving Problem
Increase in unburned carbon	Increase in stoker area Application of after-burning system
Control of furnace temperature	Flue gas recirculation Water injection Steam injection

SECTION 5

SELECTIVE CATALYTIC REDUCTION (SCR)

5.1 OVERVIEW

Selective catalytic reduction (SCR) is the most advanced flue gas treatment process for NO_x removal from waste gases. In the SCR process, NO_x is reduced to N_2 and H_2O by ammonia (NH_3) at 150-450°C in the presence of a catalyst. NH_3 is an acceptable reducing agent for NO_x in combustion gases because it selectively reacts with NO_x while other reducing agents, such as H_2 , CO , and CH_4 , readily react with O_2 in the gases. Since virtually all of the NO_x in combustion gases is in the form of NO and a small amount of O_2 promotes the reaction of NH_3 with NO , the reaction is usually expressed by the following equation. The reaction may not be exactly stoichiometric.



SCR usually removes 60-85% of NO_x using 0.61-0.90 mol NH_3 for 1 mol NO_x and leaves 1-5 ppm unreacted NH_3 . Addition of a larger amount of NH_3 increases the NO_x removal efficiency but produces a larger amount of unreacted NH_3 . The optimum temperature for SCR is 300-400°C. Therefore, SCR is conveniently applied to the 300-400°C flue gas at the boiler economizer outlet. At lower temperatures, the reaction rate is slower, while higher temperatures adversely affect catalytic activity. Low-temperature catalysts for use at 150-200°C and high temperature catalysts for use at 500-550°C also have been developed for special gases.

In the early stages of its development, SCR had the following problems:

- 1) catalyst poisoning by SO_x in the gas; 2) plugging of the catalyst by dust;
- 3) ammonium bisulfate deposition on the catalyst below about 300°C ; 4) ammonium bisulfate deposition in the air preheater below about 250°C ; 5) catalytic promotion of oxidation of SO_2 to SO_3 ; and 6) erosion of the catalyst by fly ash from coal.

Those problems have been solved by the following countermeasures:

- o Use of base metal catalysts with TiO_2 instead of Al_2O_3 or Fe_2O_3 substrates;
- o Use of parallel-flow type catalysts such as honeycomb, plate, and tube catalysts;
- o Maintaining the gas temperature above 330°C by using an economizer by-pass system;
- o Keeping unreacted NH_3 (NH_3 at the reactor outlet) below about 3 ppm;
- o Using a low-oxidation catalyst; and
- o Using a moderate gas velocity, a hard catalyst, and a device for erosion prevention such as dummy spacer.

As a result of these improvements, SCR has become highly reliable and less costly. It has been used in over 160 commercial plants with a total capacity of 27,000 MW by the end of 1984.

Major SCR plant constructors and the number and capacity of the plants constructed by them are shown in Table 5-1. Nearly all of the plants for utility boilers have been constructed since 1979 while virtually all of the plants for industrial use were constructed before 1978. Some of the old

TABLE 5-1. NUMBER AND CAPACITY OF MAJOR SCR PLANTS

Plant Constructor	Utility boiler ^a		Other sources ^a	
	Number	Capacity ^b	Number	Capacity ^b
Asahi Glass			1	70
Hitachi Ltd. & Babcock Hitachi	21	19,540	8	1,630
Hitachi Zosen			9	1,867
Ishikawajima Harima H.I.	15	18,505	1	200
Japan Gasoline (JGC)			5	393
Kawasaki H.I.	2 ^c	750	1	25
Kobe Steel			1	101
Kurabo Engineering			1	30
Mitsubishi H.I.	32	25,950	7	713
Mitsubishi Kakoki			10	244
Mitsui Toatsu Chemical			13 ^d	1,297
Mitsui Engineering			12	1,320
Sumitomo Chemical			6	999
Sumitomo Chemical Engineering			6	130
Total	70	64,754	82	9,792

^aBy the end of 1983.^bCapacity: 1000 Nm³/hr^cOne of the plants is in the U.S.^dTogether with Techno Universe Co. Five of the plants (total capacity 350,000 Nm³h) are at nitrophosphate factories in the U.S.S.R.

plants for industrial use experienced the problems described above; however, nearly all of the plants constructed since 1979 have been operated in an automatic mode without problems. Since 1980, most of coal-fired utility boilers have incorporated both SCR and limestone-gypsum FGD.

5.2 SCR CATALYSTS

5.2.1 Catalyst Shapes

Between 1975 and 1978, SCR was applied mainly to clean or semi-dirty gases from natural gas- or low-sulfur oil-fired industrial boilers and furnaces. A fixed-bed pellet catalyst was mainly used for the cleaner applications, while for a dirty gas rich in particulates, a pellet catalyst in a moving bed was often used. Parallel-flow type catalysts have been developed since 1978, and have been applied to many utility boilers (Table 5-2).

Cross-sectional shapes of the major parallel-flow type catalysts are shown in Figure 5-1. Virtually all the particulates in flue gases pass through the channels of the parallel-flow catalysts, but a small amount of very fine particulates are deposited in corners where there are sharp angles. From this standpoint, the hexagonal honeycomb (angle 120°) is superior to the triangle honeycomb (angle 60°). Actually the square honeycomb is more popular than the hexagonal honeycomb because of its greater mechanical strength.

5.2.2 Catalyst Composition and Properties

The composition and properties of parallel-flow type catalysts are compared in Table 5-3. The more popular catalysts are the ceramic honeycomb, the molded (square) or coated (hexagonal), and base-metal coated ones. The most popular sizes have a wall thickness of 0.6-1.8 mm and a channel diameter of 4-9 mm.

Most catalysts for SO_2 -containing gas consist of TiO_2 with a small amount of V_2O_5 . The combination is very active although V_2O_5 promotes the oxidation

TABLE 5-2. NUMBER AND CAPACITY OF SCR UNITS FOR UTILITY BOILERS
(Completed by the end of 1983)

Fuel	Catalyst Type	Number of Units	Total Capacity (MW)
Coal	Honeycomb	8	1,993
	Plate	6	1,361
	Tube	1	125
Oil	Honeycomb	38	14,450
	Plate	12	3,860
Gas	Pellet	6	3,900
Total		71	25,689

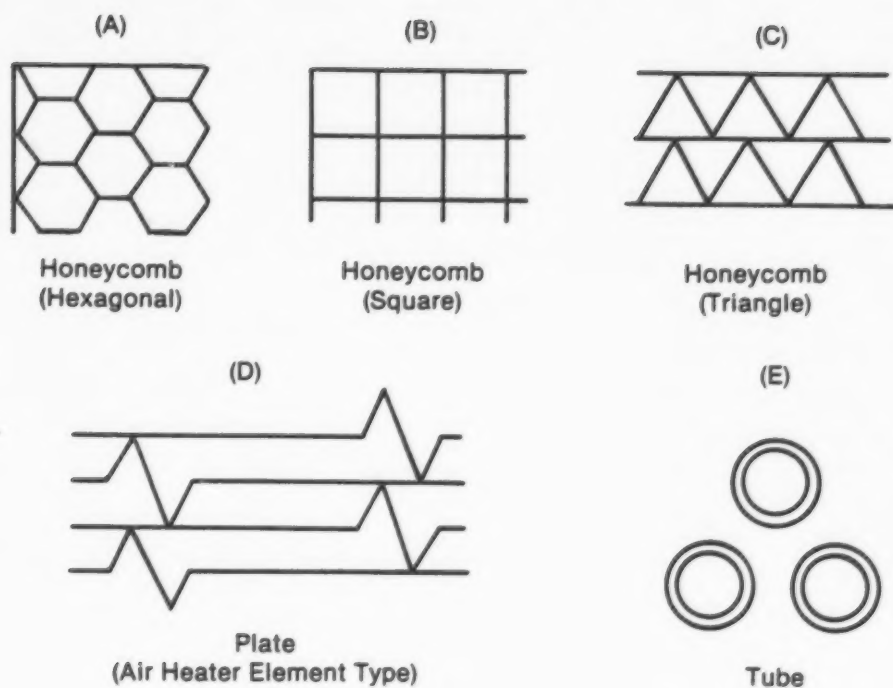
TABLE 5-3. COMPARISON OF PARALLEL-FLOW TYPE CATALYSTS

Composition	Shape No. (from Fig. 5-1)	Simplicity of Production	Mechanical Strength	Variety of Composition
Ceramic				
Solid (molded)	B, E	x	†	x
Ceramic carrier, coated	A, B	0	0	x
Ceramic carrier, impregnated	A, B	0	0	0
Metal base				
Metal plate, coated	D	†	x	x
Metal gauze, coated	C	0	0	x
Metal plate, etched	C	0	x	†

x = Superior

0 = Medium

† = Inferior



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Figure 5-1. Cross section of parallel flow catalysts.

of SO_2 to SO_3 . Therefore, to produce a low-oxidation catalyst, V_2O_5 must be reduced or eliminated and other metal oxides, such as WO_3 or MoO_3 , must be added (10).

5.2.3 Space Velocity (SV) and Area Velocity (AV)

In order to express the amount of catalyst required, the concept of "space velocity" (SV) or, for parallel-flow type catalysts, "area velocity" (AV) is used. These are defined as follows:

$$\text{SV (h}^{-1}\text{)} = \frac{\text{Volume of gas treated (Nm}^3\text{/h)}}{\text{Catalyst volume (m}^3\text{)}}$$

SV is positively affected by a more reactive catalyst, or one with a smaller pitch.

$$\text{AV (m/h)} = \frac{\text{Volume of gas treated (Nm}^3\text{/h)}}{\text{Apparent surface area of the catalyst (m}^2\text{)}}$$

Apparent surface area is the surface area assuming that the catalyst surface is smooth, without pores. Examples of the relationship between AV and SV for a square honeycomb catalyst are shown in Table 5-4.

TABLE 5-4. AV AND SV FOR A SQUARE HONEYCOMB CATALYST

Channel Diameter (mm)	Wall Thickness (mm)	Pitch (mm)	AV ₋₁ (m/h ⁻¹)	SV ₋₁ (h ⁻¹)
5	1	6	5	2,800
			10	5,600
7	1.5	8.5	5	1,900
			10	3,800
10	2	12	5	1,400
			10	2,800

Since there are parallel-flow type catalysts of various pitches, catalyst efficiency is better expressed by AV than SV. On the other hand, catalyst cost is better shown by SV because the cost (in Japan) is usually expressed by yen/m³ and most parallel-flow catalysts with different pitches currently cost about 3.0-3.5 million yen/m³.

5.2.4 Recent Trends in SCR Catalysts

Some of the recent trends in SCR catalyst use are as follows:

- o Use of low-oxidation catalysts with an oxidation ratio of 0.5% or even smaller, particularly for SO₂-rich gas.
- o Use of parallel-flow type catalysts, even for clean gas, since this catalyst type causes less pressure drop than pellet catalysts (see Section 5.4).
- o Use of catalysts with a small pitch to increase the efficiency and to reduce cost. For example, for flue gas from a coal-fired boiler with full dust load, a 10 mm pitch catalyst was used in 1980-1981, while a smaller amount of a 6-7 mm pitch catalyst has been used since 1982.

5.2.5 Disposal of Spent Catalyst

So far there has been little production of spent catalysts because catalyst life has been much longer than expected. Nearly all of the original catalysts are still in use. The longest period for continuous commercial catalyst use thus far is 9 years for clean gas (natural gas burning), 6 years for semi-dirty gas (heavy oil burning), and 3 years for flue gas from coal with full dust load. When a catalyst is degraded and has to be replaced, the catalyst producer will reclaim all of the spent catalyst since its composition is proprietary. Heavy metal components will be recovered and the remaining TiO₂ component will be sold to a TiO₂ manufacturer.

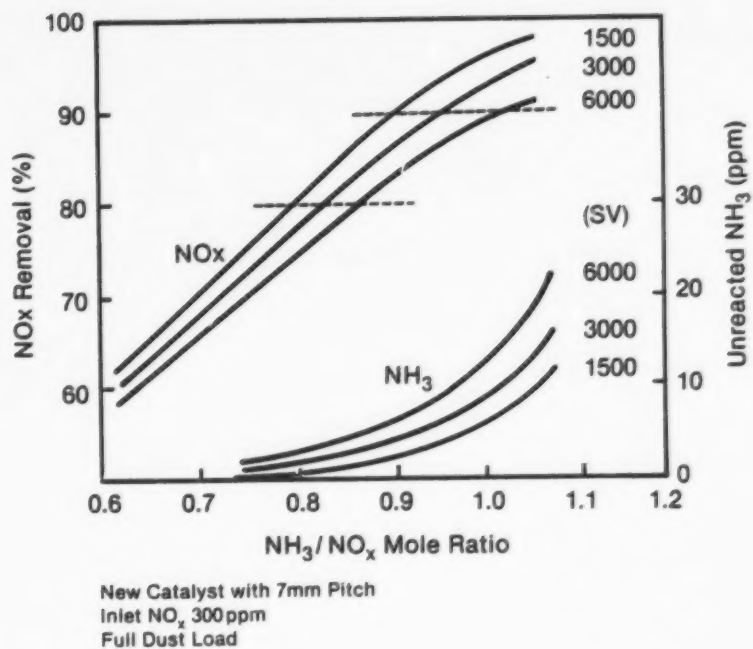
5.3 SCR PERFORMANCE

5.3.1 NO_x Removal and Unreacted NH_3

Figure 5-2 shows an example of SCR for a coal-fired boiler flue gas using a 7 mm pitch honeycomb catalyst. NO_x removal efficiencies of 80 and 90% may be attained under the conditions shown in Table 5-5. Since it is advantageous to maintain the concentration of unreacted NH_3 below 3 ppm to prevent such problems as air preheater plugging and fly ash contamination, 80% NO_x removal may be achieved with an SV of 3,000 h^{-1} . Ninety percent removal may be achieved with an SV of about 1,500 h^{-1} . Actually 90% removal with a low concentration of unreacted NH_3 is not easily attained for large amounts of flue gas such as that from large utility boilers. This is because gas velocities and NO_x concentrations may differ somewhat from place to place in a large duct. Also, changes in gas volume and NO_x concentration due to boiler load fluctuation may cause temporary increases in NO_x or NH_3 emissions. This could occur because the ammonia feed rate is controlled by sampling the flue gas volume and inlet NO_x concentration. A few minutes may be required for this analysis and the subsequent adjustment by the automatic control system.

TABLE 5-5. EXAMPLE OF NO_x REMOVAL CONDITIONS
(Tabulated from Figure 5-2)

80% NO_x Removal			90% NO_x Removal		
SV (h^{-1})	NH_3/NO_x Mole Ratio	Unreacted NH_3 (ppm)	SV (h^{-1})	NH_3/NO_x Mole Ratio	Unreacted NH_3 (ppm)
1500	0.81	1	1500	0.92	2
3000	0.83	3	3000	0.97	7
6000	0.87	7	6000	1.04	18



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Figure 5-2. SCR for coal-fired boiler flue gas at 350°C using a honeycomb catalyst.

5.3.2 Inlet NO_x Concentration and SV

Figure 5-3 shows the relative amounts of catalyst required for different NO_x removal efficiencies. Figure 5-4 shows the relative amounts of catalyst required for different inlet NO_x concentrations. A larger amount of catalyst is needed, not only for higher NO_x removal efficiencies, but also for higher inlet NO_x concentrations, provided that the concentration of unreacted NH₃ is kept the same. Therefore, it is usually more economical to reduce the inlet NO_x concentration by combustion modification.

5.3.3 Degradation of Catalyst Activity

Catalyst activity decreases slowly during flue gas treatment. Degradation is affected by the amount of impurities such as SO₃ and alkaline compounds in the gas, as well as the composition of the catalyst, and the temperature. Figure 5-5 shows a typical example for flue gas from boilers using different fuels at 350-400°C. Assuming the conditions as shown in Figure 5-5, the catalyst life for a NO_x removal efficiency of 80% may be about 3 years for coal-, 5 years for low-sulfur oil-, and over 5 years for gas-fired boilers. Actual operation of boilers in SCR plants verifies these results.

Catalysts may be seriously contaminated by a special gas such as that from a glass melting furnace which may contain a considerable amount of vaporized sodium compounds. Sodium compounds will condense even on parallel-flow catalysts. In such cases, the catalyst has to be occasionally washed with water to remove the sodium compounds.

When a catalyst is used with a low-temperature gas (below 300°C) containing SO_x, ammonium bisulfate (NH₄HSO₄) tends to deposit on the catalyst and lowers its activity. In this case, the catalyst is occasionally heated to over 350°C to remove the bisulfate.

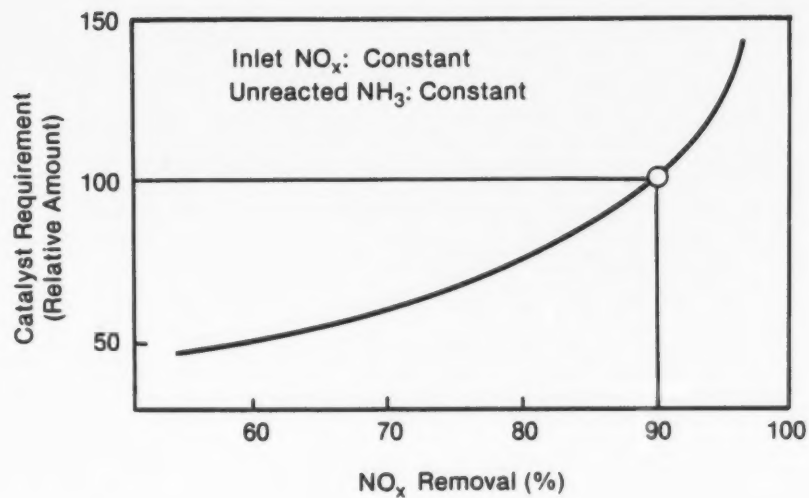


Figure 5-3. NO_x removal efficiency vs. relative amount of catalyst (by Babcock-Hitachi).

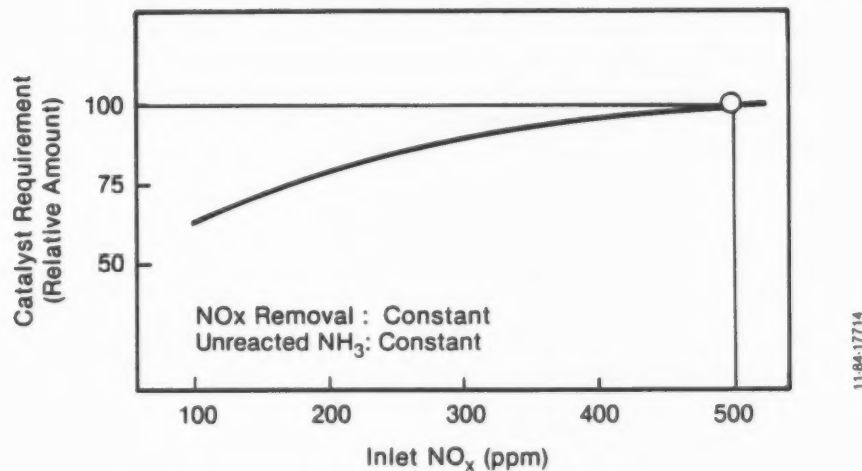


Figure 5-4. Inlet NO_x concentration vs. relative amount of catalyst (by Babcock-Hitachi).

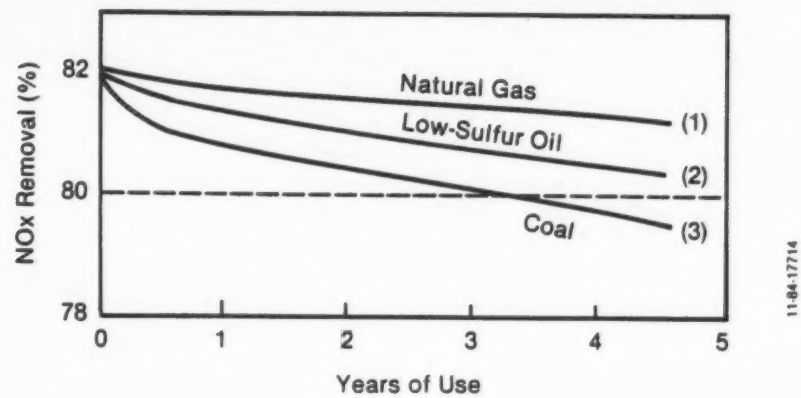


Figure 5-5. Degradation of catalyst activity in flue gases from different fuels.

5.4 SCR FOR UTILITY BOILERS

5.4.1 Status

The actual performance of major SCR plants for utility boilers is shown in Table 5-6. SCR plants with coal-fired boilers use parallel-flow catalysts with an SV of 2,000-4,000 h^{-1} to remove about 60-80% of NO_x ; those with oil-fired boilers burning low-sulfur oil use parallel-flow catalysts with an SV of more than 5,000 h^{-1} to remove 80% of NO_x . The small SV for coal is due to the high inlet NO_x concentration (refer back to Figure 5-4) and the higher SO_x concentration which necessitates the use of a less reactive, low oxidation catalyst. SCR plants with gas-fired boilers use pellet catalysts having an SV of 20,000 h^{-1} .

New SCR plants for existing utility boilers or those planned for completion between 1983 and 1987 are listed in Table 5-7. SCR plants for gas-fired boilers completed in 1983 use pellet catalysts. However, new SCR plants for gas-fired, combined-cycle power plants will use either honeycomb or plate catalysts because of the low pressure drop and the recent development of efficient, small pitch, parallel flow catalysts.

5.4.2 Boiler Load Change and SCR

Typical performance for a utility boiler SCR unit during load change is shown in Figure 5-6. When the boiler load is reduced, the gas temperature is lowered but the NO_x removal efficiency usually increases. This is because the gas volume has decreased, resulting in a smaller SV or a longer reaction time. The SO_2 oxidation ratio will usually decrease at the reduced temperature.

When the temperature is lowered, ammonium bisulfate may deposit on the catalyst. The temperature of deposit formation on the catalyst is usually 300-330°C, and about 250°C with an air preheater. Bisulfate formation on the catalyst at the higher temperature may be due to 1) the formation of a high concentration of SO_3 on the catalyst surface from the oxidation of SO_2 and 2)

TABLE 5-6. PERFORMANCE OF MAJOR SCR PLANTS ON UTILITY BOILERS

Fuel	Plant Owner	Plant Site	Boiler No.	MW	SCR Mfr.	Catalyst Type	Pitch (mm)	SV (h ⁻¹)	NO _x (ppm) In	NO _x (ppm) Out	DeNO _x (%)	Unreacted NH ₃ (ppm)	Start Up Year
EPDC	EPDC	Takehara	1	250xl/2	BHK	Plate	10	2,300	350	67	81	4	1981
		Takehara	2	250xl/2	KHI	Tube	-	2,000	350	67	81	4	1981
		Takehara	3	700	BHK	Plate	10	2,370	250	48	81	2	1983
COAL	Chugoku	Shimonoseki	1	175	MHI	Honeycomb	10	3,000	420	180	57	1	1980
		Shin-Ube	3	156	MHI	Honeycomb	7	4,000	400	140	65	2	1981
		Mizushima	2	156	BHK	Plate	6	2,200	350	120	65	2	1984
		Saijo	1	156	MHI	Honeycomb	7	4,000	380	130	65	1	1983
		Saijo	2	250	IHI	Honeycomb	7.5	-	330	90	70	1	1983
OIL (Low S)	Chugoku Chugoku Chubu	Kudamatsu	2	375	IHI	Honeycomb	-	5,500	150	28	81	5	1979
		Kudamatsu	3	700	IHI	Honeycomb	-	5,500	110	21	81	6	1979
		Chita	4	700	MHI	Honeycomb	8	5,430	100	18	82	2	1979
		Chita	5, 6	700	BHK	Pellet	5	20,000	40	8	80	4	1975
GAS	Chubu	Chita II	1	700	BHK	Pellet	5	20,000	50	10	80	4	1983

TABLE 5-7. NEW SCR PLANTS ON UTILITY BOILERS
(Projected Completion 1983 - 1987)

Company	Station	Boiler			New or Retrofit	SCR Mfr.	Catalyst Type	Year Completed
		No.	Fuel	MW				
Chubu	Chita II	1	Gas	700	N	BHK ^a	Pe ^b	1983
Chubu	Chita II	2	Gas	700	N	IHI ^c	Pe	1983
Chubu	Yokkaichi	4	Gas	560 ^d	N	HZ ^e	Ho ^f	1987
Chugoku	Tamashima	1	Oil	350	R	BHK	Pl ^g	1983
Chugoku	Mizushima	1	Coal	125	R	BHK	Pl	1984
Chugoku	Mizushima	2	Coal	156	R	BHK	Pl	1984
Chugoku	Mizushima	3	Oil	350	R	BHK	Pl	1983
Chugoku	Shin Onoda	1	Coal	500	N	IHI	Ho	1986
Chugoku	Shin Onoda	2	Coal	500	N	IHI	Ho	1987
EPDC	Takehara	3	Coal	700	N	BHK	Pl	1983
Johan	Nakoso	8	Oil, Coal	600	N	MHI ^h	Ho	1983
Johan	Nakoso	9	Oil, Coal	600	N	IHI	Ho	1983
Kansai	Aioi	3	Oil	375	R	IHI	Ho	1983
Kansai	Gobo	1	Oil	600	R	MHI	Ho	1984
Kansai	Gobo	2	Oil	600	R	BHK	Pl	1984
Kansai	Gobo	3	Oil	600	R	N.D. ⁱ		1985
Kyushu	Minato	1	Coal	156	R	MHI	Ho	1983
Kyushu	Shin Kokura	5	Gas	600	N	MHI	Pe	1983
Kyushu	Matsuura	1	Coal	700	N	MHI	Ho	1987
Sakata	Sakata	1	Coal	350	N	MHI	Ho	1984
Sakata	Sakata	2	Coal	350	N	MHI	Ho	1987
Shikoku	Saijo	1	Coal	156	R	MHI	Ho	1983
Shikoku	Saijo	2	Coal	250	R	IHI	Ho	1984
Tohoku	Sendai	2	Coal	175	R	BHK	Pl	1983
Tohoku	Sendai	3	Coal	175	R	BHK	Pl	1983
Tohoku	Higashi Niigata	2	Oil	600	R	IHI	Ho	1983
Tohoku	Higashi Niigata	3	Gas	545x2 ^d	N	MHI	Ho	1984, 1985
Tokyo	Yokosuka	1	COM ^f	265	R	MHI	Ho	1984
Tokyo	Yokosuka	2	COM	265	R	BHK	Pl	1985
Tokyo	Futtsu	1	Gas	1000 ^d	N	HZ	Ho	1986
Tokyo	Futtsu	2	Gas	1000 ^d	N	HZ	Ho	1986
Toyama	Toyama	1,2	Coal	200	R	BHK	Pl	1983

^aBabcock Hitachi

^bPellet

^cIshikawajima Harima Heavy Industries

^dCombined cycle

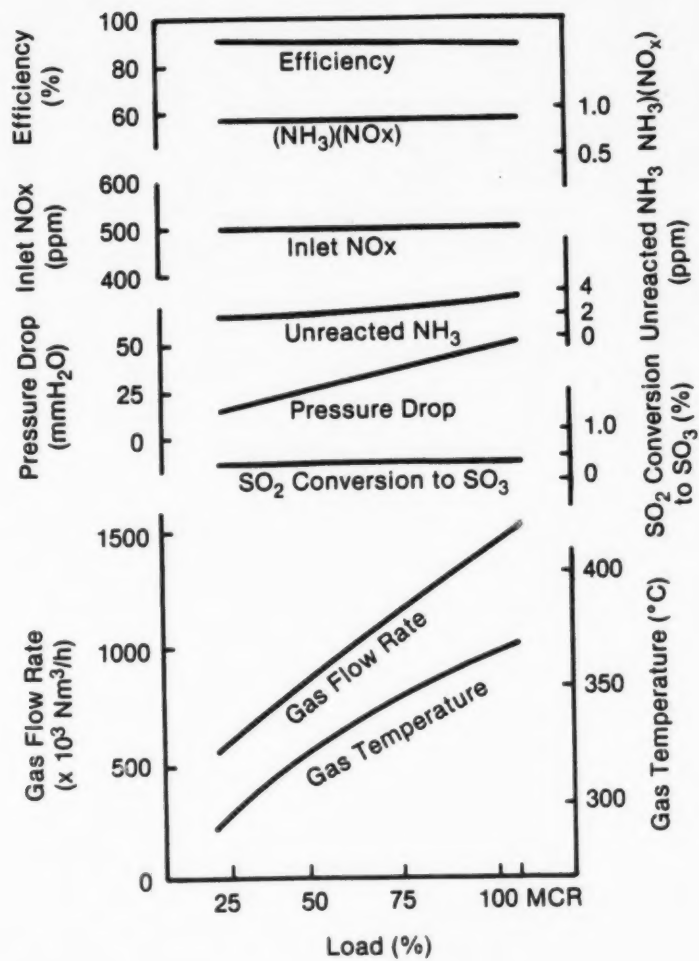
^eHitachi Zosen

^fHoneycomb

^gPlate

^hMitsubishi Heavy Industries

ⁱNot described



11-84-17714

Figure 5-6. Expected performance of an SCR unit for a utility boiler (10).

high NH_3 concentration in the reactor. Therefore, SCR plants for coal-fired boilers usually have an economizer by-pass line to maintain the gas temperature above about 320°C in the SCR reactor even at a reduced boiler load. When the by-pass is used, less heat is recovered by the economizer, but it can be recovered instead by the air preheater located downstream.

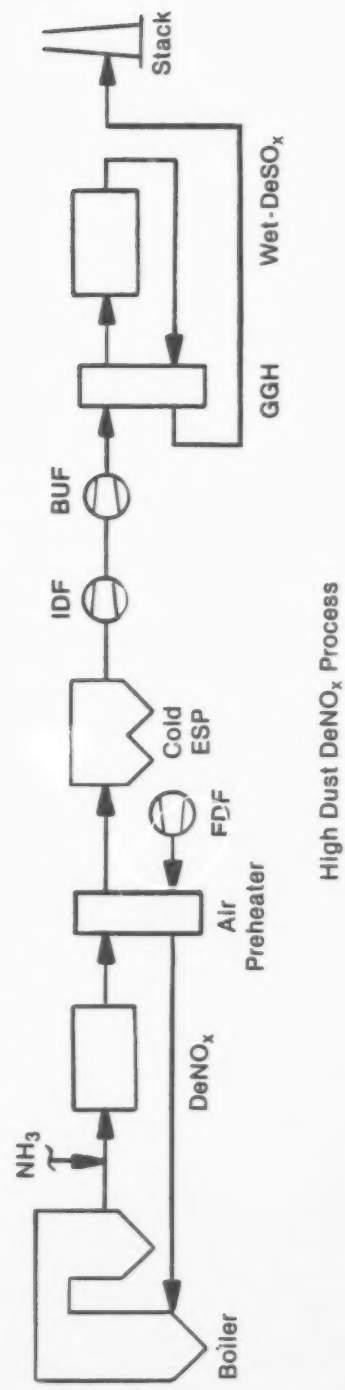
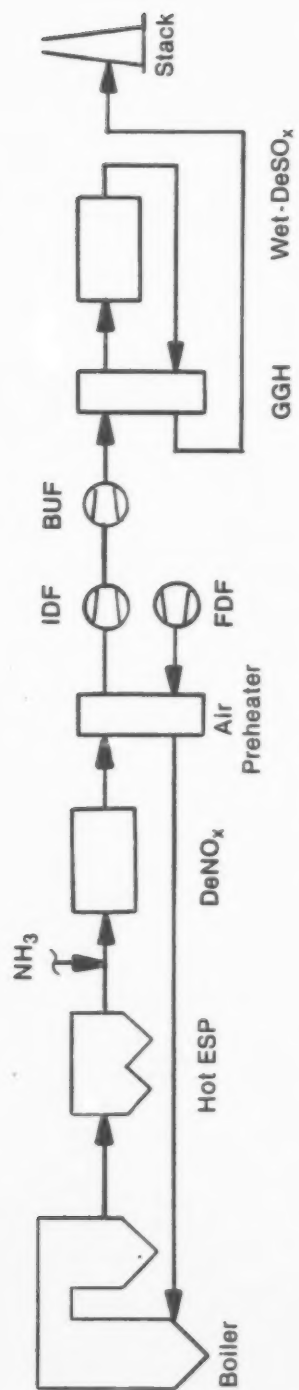
New SCR plants maintain the concentration of unreacted NH_3 below 3 ppm so that no bisulfate problem is encountered in the air preheater. SCR plants for utility boilers are usually operated automatically with no problems.

5.4.3 SCR Systems for Coal-fired Boilers

Two types of SCR systems are applied to coal-fired boilers, as shown in Figure 5-7. In the low-dust system, the flue gas is first treated in a hot-side ESP for dust removal before being subjected to SCR, while in the high-dust system, flue gas with a full dust load (about 20 g/Nm^3) is treated by SCR directly. The advantages and disadvantages of the systems are compared in Table 5-8.

TABLE 5-8. COMPARISON OF HIGH-DUST AND LOW-DUST SCR SYSTEMS FOR COAL-FIRED BOILERS

	High-Dust System	Low-Dust System
Catalyst erosion by dust	More	Less
Dust deposition in reactor	Less	More
Ammonium bisulfate deposition in air preheater	Less	More
Ammonium compound contamination of fly ash	Possible	None
Cost of ESP	Less	More
Suitability of ESP	Suitable to medium and high-sulfur coals	Suitable to low-sulfur coal



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Figure 5-7. Total flue gas treatment process for coal-fired boilers.

With the low-dust system, catalyst erosion rarely occurs, but the dust leaving the hot ESP consists of fine, relatively alkaline ash which tends to adhere to the surface of the catalyst. Ammonium bisulfate deposits also tend to form in the air preheater. With the high-dust system, most of the bisulfate deposits on the ash so that the air preheater has no ammonium bisulfate problem. Therefore, when the gas is rich in unreacted ammonia, fly ash caught by the cold ESP may present a utilization and disposal problem. However, as mentioned earlier, the problem does not usually occur when the NH_3 concentration is kept below 5 ppm. Fly ash erosion of the catalyst can be prevented if a hard catalyst and a dummy spacer are used and the gas velocity is kept moderate.

Although both low-dust and high-dust systems are feasible if the concentration of unreacted NH_3 is kept below 3 ppm, the latter system is more popular than the former, as was shown in Table 2-6.

5.4.4 SCR/FGD Combination

The combined SCR-FGD systems for coal-fired boilers are shown in Figure 5-7. The major difficulty with the combined system is the possibility of unreacted NH_3 in the flue gas being caught by the FGD system. If this NH_3 concentration is kept below 5 ppm, then the resulting concentration of ammonia in the scrubber liquor is usually below 300 mg/liter and does not affect scrubber operation. The ammonia content in the flue gas leaving the FGD plant is below 1 ppm, causing no plume problem.

A greater problem is the ammonia contained in the wastewater from the FGD plant. An unreacted NH_3 concentration of 3 ppm in the flue gas from a 500 MW boiler amounts to 4 kg/h. If the FGD plant purges 20 t/hr of wastewater, the water will contain 200 mg/liter of ammonia. Wastewater from FGD plants for coal-fired boilers contains 100-600 mg/liter of nitrogen in various forms when SCR is not applied and these nitrogen compounds are derived from NO_x in the flue gas. With SCR, ammonia increases in the wastewater but the amount of nitrogen compounds decreases because of the lowered concentration of NO_x in the flue gas. Thus the net amount of nitrogen (all forms) in the wastewater

may not be increased appreciably. If necessary, the ammonia in the wastewater can be removed by biological treatment, as has been done at EPDC's Takehara plant. However, most plants do not have ammonia removal facilities.

The major problem for the FGD system is dust plugging of the gas-gas heater. The oxidation of SO_2 to SO_3 by an SCR catalyst tends to increase deposit formation. On the other hand, the unreacted NH_3 in the gas combines with SO_3 and may reduce the effect of SO_3 . The gas-gas heater can be operated continuously for a year by using a low-oxidation catalyst and by adjusting the inlet dust concentration as described in Section 2.6.

5.5 SCR FOR COMBINED CYCLE POWER PLANTS

5.5.1 Overview

Combined cycle power generation (gas turbine and steam turbine) has become popular in Japan because combined cycle has a higher thermal efficiency (42-43%) than conventional steam turbine boilers (39-40%). For fuel, natural gas is used in large utility power station systems while oil is used for most small industrial applications.

In the combined cycle process, a gas turbine burns the fuel with a large amount of excess air. The flue gas contains approximately 15% O_2 and about three times as much flue gas is produced as from conventional boilers. (Flue gas from conventional boilers also contains much less oxygen: 1-2%). Most of the gas turbines apply steam or water injection to reduce NO_x . The injection process is fairly costly because a large amount of purified water is required and fuel consumption is increased. Also, even with steam injection, NO_x concentrations in the flue gas (15% O_2 basis) range from 35 to 70 ppm for natural gas and from 50 to 150 ppm for oil-firing. These amounts of NO_x are much larger than that which is emitted from conventional boilers. Since further NO_x abatement is required by local governments for most of the new combined cycle plants, SCR is being applied to remove 60-85% of the NO_x .

Japanese combined cycle plants with SCR are listed in Table 5-9. The Kawasaki plant is owned by the Japanese national railway and has been operated for 3 years. Tohoku Electric's Higashi Niigata plant, which began operation

TABLE 5-9. COMBINED CYCLE POWER PLANTS WITH SCR IN JAPAN

Owner	Plant Site	Boiler Number	Fuel	Capacity (MW)	SCR Maker	Start-up Year (Actual or Projected)
Japan National Railway	Kawasaki	1	Kerosene	144	BHK ^a	1981
Tohoku Electric Power	Higashi Niigata	3	LNG	545	MHI ^b	1984
				545	MHI	1985
Tokyo Electric Power	Futtsu	1	LNG	1,000	HZ ^c	1986
		2	LNG	1,000	HZ	1986
Chubu Electric Power	Yokkaichi	4	LNG	560	HZ	1987
Kyushu Electric Power	Shin-Oita	1	LNG	690	BHK	1989
		2	LNG	870	MHI	1989

^aBabcock-Hitschi K.K.^bMitsubishi Heavy Industries^cHitachi Zosen

in 1984, does not apply steam injection and reduces NO_x to 15 ppm (15% O_2 basis) with the combination of a low- NO_x combustor and SCR. The Futtsu plant of the Tokyo Electric Power Co. is currently under construction and is projected to be on-line in 1986. These three plants are described in more detail in the following three sections.

In a combined cycle plant with SCR, the flue gas temperature at the gas turbine outlet is above 500°C. The gas is cooled by a superheater and a pre-evaporator to 280-400°C and is then subjected to SCR. After the SCR reactor, the gas is passed through an evaporator and an economizer for heat recovery.

SCR for combined cycle applications requires a large amount of catalyst because of the large gas volume. Also, the catalyst must be resistant to thermal shock because the gas temperature may change rapidly due to changes in the operating load. On the other hand, a highly reactive, efficient catalyst can be used because the flue gas is clean or nearly clean with little or no SO_x and particulates.

5.5.2 Kawasaki Plant, Japan National Railway (JNR) (21)

The Kawasaki Power Station generates power for electric trains. In addition to three conventional boilers, a Hitachi-General Electric combined cycle plant has been installed. This plant has been in operation since 1981. It burns kerosene (0.05% sulfur) for 14-16 hours a day, 6 days a week, with shutdown at night and on Sundays. The power generation capacity is 141.3 MW, with 97.1 MW by gas turbine and 44.2 MW by steam turbine (Figure 5-8). Flue gas volume is about 1,100,000 Nm^3/h and contains about 60 ppm NO_x (15% O_2 basis) at full load.

The SCR plant at Kawasaki was constructed by Babcock-Hitachi K.K. (BHK). The SCR reactor contains a pellet catalyst in an intermittent moving bed; gas passes horizontally through this bed.

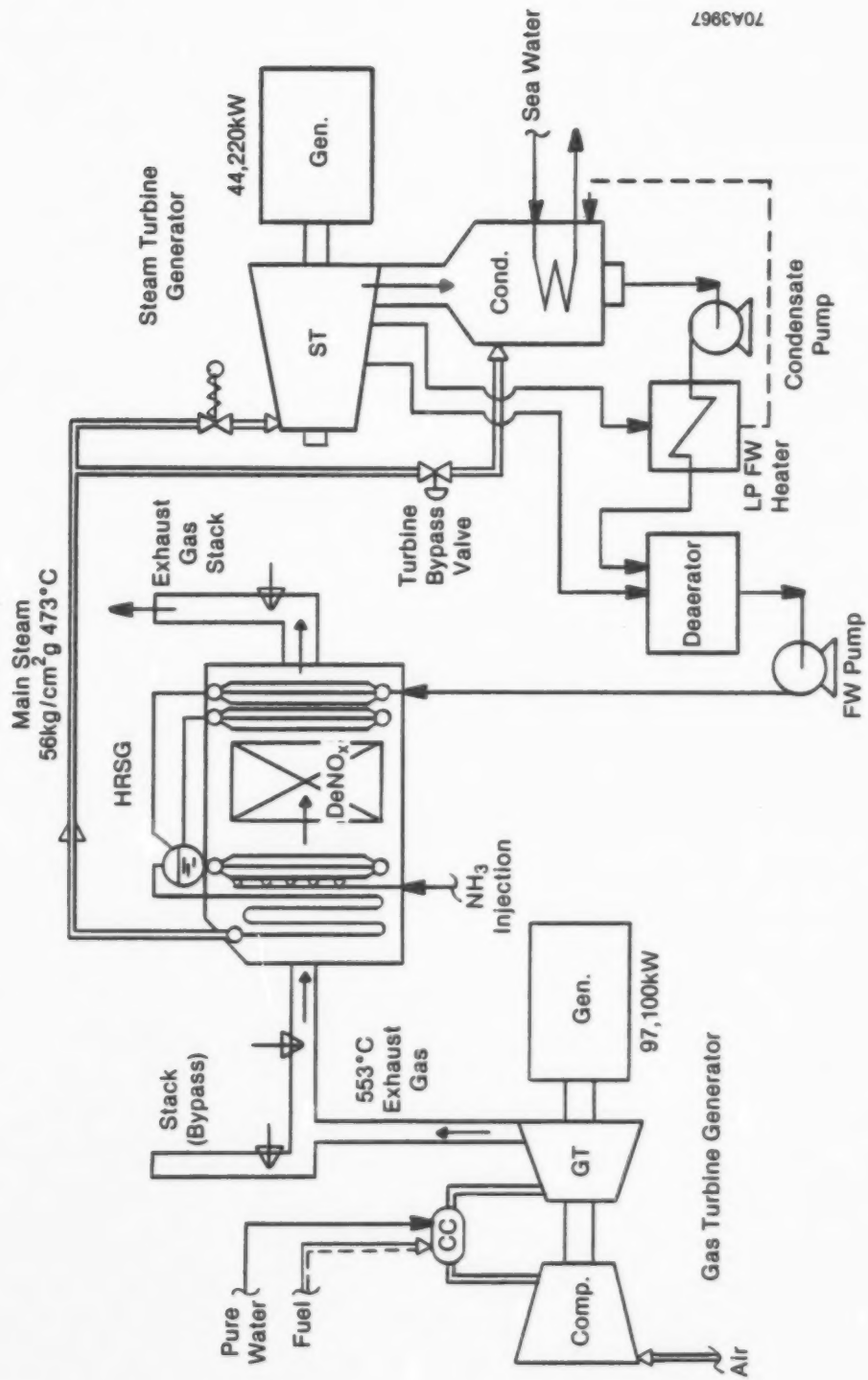


Figure 5-8. Combined cycle plant flow diagram (JNR Kawasaki Plant).

Ammonia is injected when the gas temperature is above 250°C, which is reached in 20-30 minutes after startup. Normally a NH_3/NO_x mole ratio of 0.7-0.8 is used to remove 65-75% of NO_x . This maintains NO_x below 15 ppm at 15% O_2 and unreacted NH_3 below about 2 ppm. No ammonium bisulfate problem has been experienced because of the small amount of unreacted NH_3 and low SO_x concentrations (the latter is about 10 ppm, including 1 ppm SO_3).

The Kawasaki plant has been operated since 1981 without problems. Figure 5-9 summarizes the performance of the plant. The total time of operation with SCR (ammonia injection hours) exceeded 11,000 hours by June 1984. Catalyst activity has decreased slightly during plant operation: with the NH_3/NO_x mole ratio of 0.85, the NO_x removal efficiency was about 83% initially and has now decreased to 81-82%. However, the catalyst still may be useful for three to four more years.

Hitachi Ltd. has recently developed a plate catalyst for coal-fired boiler flue gas. This plate catalyst is preferable to the pellet catalyst because its pressure drop is lower (below 100 mm H_2O) than that of the pellet catalyst in the moving bed (150-200 mm H_2O).

5.5.3 Higashi Niigata Plant, Tohoku Electric (22)

MHI has been constructing a 1090 MW combined cycle power plant at Tohoku Electric's Higashi Niigata Power Station. The plant uses LNG for fuel and consists of 6 gas turbines (118 MW x 6), 6 heat recovery steam generators, and 2 steam turbines (191 MW x 2), as shown in Figure 5-10. Half of the entire plant, including one steam turbine, 3 trains of gas turbines, and one steam generator and the SCR unit were complete in 1984, with the rest scheduled for completion in 1985.

The power plant is designed for base load operation. The flue gas from each gas turbine amounts to 1,365,000 Nm^3/h and contains about 14% O_2 . The gas turbines utilize the Premix Combustion System developed by MHI to reduce NO_x to about 100 ppm (15% O_2 basis) without steam injection. (By comparison,

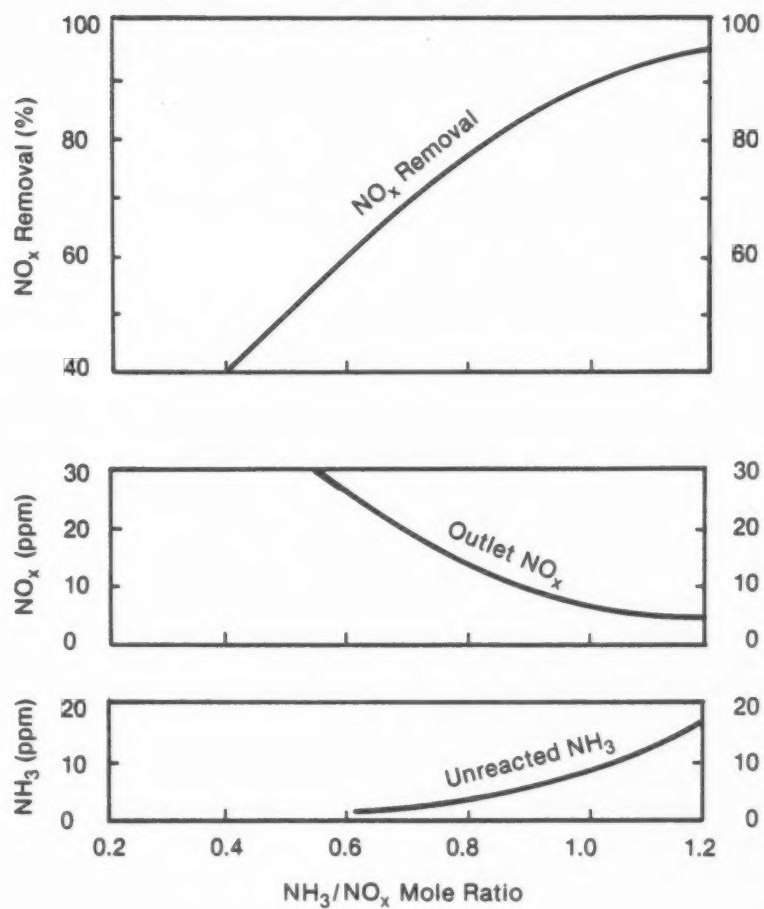


Figure 5-9. Performance of the JNR Kawasaki Plant (Inlet NO_x = 55-65 ppm at full load).

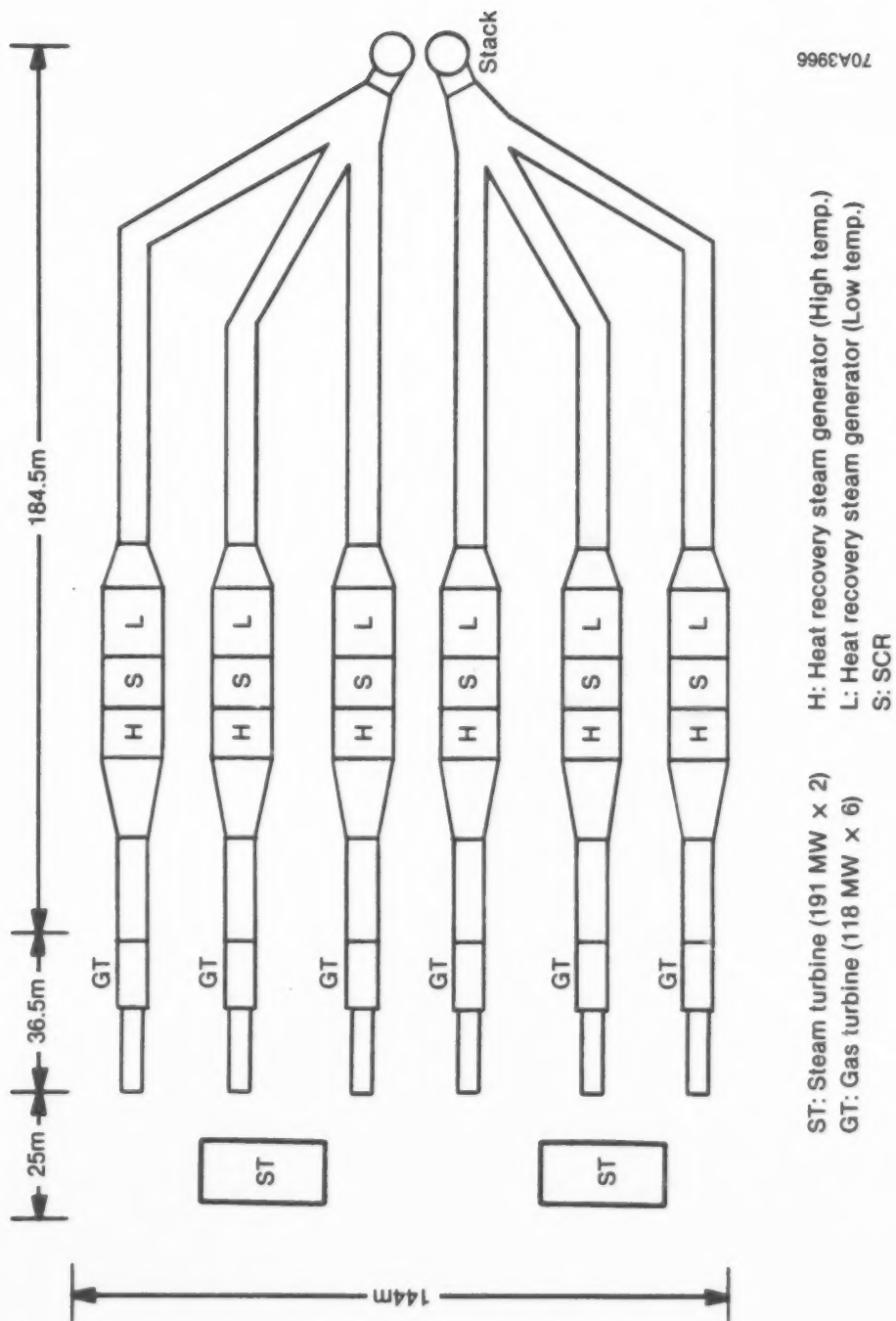


Figure 5-10. Combined cycle power plant (Higashi Niigata Power Station of Tohoku Electric, 1090 MW).

conventional combustors for gas turbines produce NO_x concentrations of about 180 ppm without steam injection). SCR is applied to lower NO_x to 15 ppm at 15% O_2 (40 ppm at 5% O_2 basis).

The SCR catalyst is placed between the high and medium temperature modules of the heat recovery steam generator. The gas at 297°C is passed horizontally through the catalyst bed. A square honeycomb catalyst with a pitch of 4.5 mm is used at an SV of $9,000 \text{ h}^{-1}$. An NH_3/NO_x mole ratio of nearly 1.0 will be used to maintain the concentration of unreacted NH_3 below 4 ppm.

The catalyst bed is 12.8 m wide and 11.8 m high. This size was determined by the size of the heat recovery steam generator. The gas velocity is 8 m/sec and the pressure drop is guaranteed to be below 140 mm H_2O . The pressure drop could also be reduced by enlarging the cross section of the catalyst bed to reduce the gas velocity. A slightly larger amount of catalyst might be needed in this case to make sure the NO_x removal efficiency is not diminished.

5.5.4 Futtsu Plant, Tokyo Electric (23, 24)

Two 1,000 MW combined cycle plants are under construction at Tokyo Electric's Futtsu Power Station. One of the plants will begin operation in 1986, the other in 1988. LNG will be used as the fuel. Each 1,000 MW plant consists of 7 units, each of which is equipped with a General Electric gas turbine and steam turbine. The unit capacity is 142.8 MW; about two-thirds of this is generated by the gas turbine and the rest by the steam turbine (Figure 5-11). The flue gas volume reaches $1,250,000 \text{ Nm}^3/\text{h}$ for each unit.

The gas turbine uses steam for NO_x abatement. The NO_x concentration is expected to be lowered to 35-40 ppm at 15% O_2 basis with the maximum steam injection, and will be reduced to 3-4 ppm by SCR using a NH_3/NO_x mole ratio of 1. Hitachi Zosen's NOXNON 500, an 8 mm pitch base-metal, honeycomb-type catalyst with an SV of $5,000 \text{ h}^{-1}$, ($\text{AV } 8.9 \text{ m h}^{-1}$) will be used. It is expected

that with the catalyst, 90% NO_x removal will be attained even after 35,000 hours of operation. The catalyst is placed between the evaporators in the heat recovery steam generator in a location with a cross section of 3 x 14 m. The flue gas flows up through the catalyst with a velocity of 10 m/sec. The pressure drop is expected to be below 150 mm H_2O at full load. The unreacted NH_3 concentration will be maintained below the outlet NO_x concentration (3-4 ppm).

5.6 SCR FOR VARIOUS GASES

5.6.1 Overview

About 50 SCR plants are used with oil- or gas-fired industrial boilers. Their performance is similar to that of SCR plants for utility boilers, except that most of the former remove about 90% of the NO_x using an equal molar NH_3/NO_x ratio and producing 5-10 ppm of unreacted NH_3 . The high unreacted NH_3 concentration usually does not hinder plant operation because the gas contains little SO_x or has a temperature above 250°C at discharge so that there is no ammonium bisulfate problem.

In addition, there are nearly 30 SCR plants treating gases with different compositions and temperatures, such as the gas from glass melting furnaces, sintering machines, coke ovens, diesel engines, incinerators, etc. SCR catalysts can treat gases rich in SO_x and dust, but may be poisoned by alkaline compounds, particularly potassium compounds, and by fluorides. For example, the gas from a glass melting furnace is rich in sodium and potassium compounds which condense as they cool in various locations, including the catalyst surface, even if it is of the parallel-flow type. One way to treat the gas is to remove the alkaline compounds by water scrubbing prior to SCR. However, costly gas reheating is then needed. Another way is to use an alkali-resistant catalyst and wash the catalyst occasionally with water to remove the alkaline compounds. This process can be difficult, however. Therefore, the usual conclusion is that gas from a glass melting furnace cannot be conveniently treated by SCR.

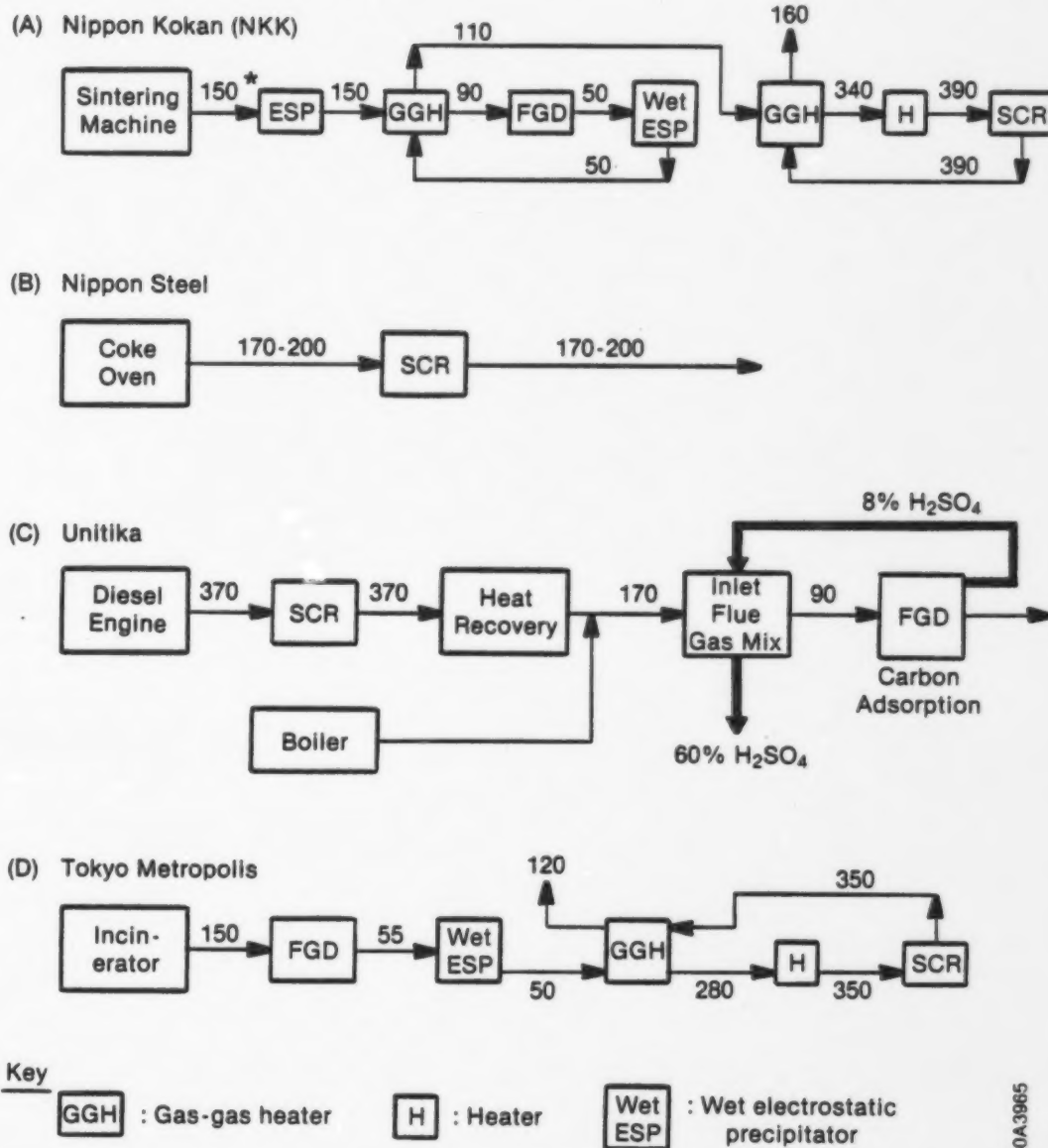
Fluoride-rich gas, such as that from pickling or from a nitrophosphate plant, is also sometimes problematic but can be treated. At Nihon Yakin's Kawasaki plant, the pickling waste gas is water-washed to remove fluorine, then heated and subjected to SCR because there is not a large amount of gas. The system was constructed by Babcock Hitachi. Techno Universe Co. constructed five SCR units in the U.S.S.R. to treat a total of 350,000 Nm³/h of fluoride-containing gas from nitrophosphate plants. The SCR units use a fluoride-resistant catalyst manufactured by Mitsui Toatsu Chemical.

A low-temperature gas (below 250°C), such as the gas from iron-ore sintering machines and coke ovens, may be treated with a low-temperature catalyst. When the gas contains SO_x, ammonium bisulfate deposits on the catalyst, necessitating the occasional heating of the catalyst above 350°C to remove the bisulfate. For a gas rich in SO_x, frequent heating would be required which is not practical. Therefore, it is necessary to remove SO_x first or to heat the gas above 350°C before SCR.

Four examples of SCR plants for various gases, including diesel engine exhaust gas and incinerator waste gas, are described in the following subsections and are summarized in Table 5-10.

5.6.2 Iron Ore Sintering Machine Flue Gas

The flue gas from iron ore sintering machines is not easily treated by SCR because of its low temperature (about 150°C) and the considerable amounts of SO_x and dust which are present. At Nippon Kokan's Keihin plant, the flue gas is first subjected to ammonia scrubbing, utilizing ammonia in coke oven gas (see Table 5-10 and item A of Figure 5-12). The gas is further treated by a wet ESP to remove particulates, and prevent any dust plugging problems in the gas-gas heater. The cleaned gas is then heated to 390°C by two more heaters and passed through an SCR system which uses an iron ore catalyst in a moving bed. Iron ore reacts with the remaining SO_x in the flue gas to form iron sulfate, which is separated from the ore by screening and then used as a feed to the sintering machine.



*Numbers Indicate Gas Temperature, °C

Figure 5-12. Systems for treatment of various flue gases.

TABLE 5-10. SCR FOR VARIOUS GASES

Owner	Nippon Kokan	Nippon Steel	Unitika	Tokyo Metropolis
Plant site	Keihin	Kimitsu	Uji	Sunamachi
Gas source	Sintering machine	Coke oven	Diesel engine	City refuse incinerator
Gas treated Nm ³ /h	1,320,000	152,000	70,000	100,000
Inlet Conc.				
SO ₂ , ppm	300	15	600	100
NO _x , ppm	200	400	1,000	100
Dust, mg/Nm ³	500	30	150	100
FGD mfr.	Nippon Kokan	-	Unitika	-
FGD process	Ammonia scrubbing	-	Carbon adsorption	Sodium scrubbing
SCR mfr.	Nippon Kokan	JGC Corp.	Unitika	Mitsui Engineering
Catalyst type	Pellet (iron ore)	Pellet (pp) ^a	Pellet	Honeycomb ^b
Reaction temp., °C	390	170-200	350-400	350
Outlet conc.				
SO ₂ , ppm	10	15	180	5
NO _x , ppm	60	40-100	150	10
Dust, mg/Nm ³	5	30	50	10

^aParallel passage reactor^bOffensive odor is also removed

Nippon Kokan first constructed the FGD unit with the wet ESP and later added the heating system and SCR unit. Therefore the system is more complex than usual. However, the process is unique in utilizing 1) ammonia in coke oven gas for SO_x scrubbing and 2) iron ore for the SCR catalyst. The plant has been operated without problems.

5.6.3 Coke Oven Gas

Nippon Steel's Kimitsu plant (see Table 5-10 and item B of Figure 5-12) treats coke oven gas at 170-200°C with a low-temperature small pellet catalyst developed by JGC, in a parallel passage reactor developed by Shell. Since ammonium bisulfate deposits on the catalyst, the catalyst is occasionally heated above 350°C to remove the bisulfate. The frequency of the heating increases with the SO_x content of the gas; the process may be too costly for a gas rich in SO_x .

5.6.4 Diesel Engine Exhaust Gas

Unitika Co. constructed an SCR unit with a gas treatment capacity of 70,000 Nm^3/h for the exhaust gas from a diesel engine which contains about 1,000 ppm NO_x , 600 ppm SO_x , and 100-150 mg/Nm^3 dust with 13% O_2 . The SCR unit uses a spherical, 4 mm TiO_2 -based catalyst (SV of 10,000 h^{-1}) in an intermittent moving bed. Approximately 10% of the catalyst is replaced annually. About 85% of the NO_x is removed by the SCR unit, using 0.85 mol NH_3 to 1 mol NO_x . The plant has been operated for 6 years without problems.

The flue gas leaving the SCR reactor is subjected to heat recovery and combined with the flue gas from an oil-fired boiler. The combined gas--170,000 Nm^3/h containing 1,500 ppm SO_2 --is subjected to activated carbon adsorption to remove nearly 90% of the SO_2 (see item C in Figure 5-12). The carbon is washed with water to recover weak H_2SO_4 (about 8%) which is sprayed into the inlet flue gas at 170°C to concentrate the acid to 60%. The recovered acid is used at Unitika's chemical plant. The FGD plant has been operated for nearly 10 years without significant problems.

5.6.5 Incinerator Flue Gas

In 1979, Mitsui Engineering constructed the first unit to remove both NO_x and odor from a sewage sludge incinerator that produces $100,000 \text{ Nm}^3/\text{h}$ of flue gas. The gas contains more than 100 ppm NO_x with considerable amounts of SO_x , HCl , various kinds of dust, and odorous components and is treated by sodium scrubbing and wet ESP (see Table 5-10 and item D of Figure 5-12). The treated gas contains about 100 ppm NO_x , 5 ppm each of SO_x and HCl with odorous components, and less than 10 mg/Nm^3 of dust. The gas at $40\text{--}50^\circ\text{C}$ is heated to 350°C in two stages, injected with NH_3 , and sent to a reactor containing a honeycomb catalyst with an SV of $4,000 \text{ h}^{-1}$. Over 90% of NO_x and 80% of the odor are removed by using 1.1 mol NH_3 to 1 mol NO_x . The plant has been operated successfully since the system start-up.

Mitsui recently constructed more than 10 similar units for incinerators. All of the incinerators had existing wet FGD plants. In each case, SCR has been applied downstream from the FGD unit after gas heating. Since gas heating is costly, Mitsui has also developed a catalyst which is resistant to HCl and is usable with flue gas before scrubbing. The company is planning to install a flue gas cleaning system, including SCR and HCl removal, for city refuse incinerators. In this system, SCR will be applied directly to hot incinerator gas which will then be subjected to HCl removal.

5.7 SCR COST

The estimated costs of SCR for utility power plants are shown in Table 5-11. These estimates are made for 1) conventional coal-, oil-, and gas-fired 700 MW boilers with 2 SCR reactors each, 2) 175 MW wet bottom coal-fired boilers with one SCR reactor, and 3) 700 MW gas-fired combined cycle plants in five trains, each with an SCR reactor. An NO_x removal ratio of 80% was assumed except for the wet bottom boiler with a high inlet NO_x concentration (1,000 ppm) for which both 80% and 90% levels of removal were assumed. Unreacted ammonia was assumed to be present in low concentrations, 2-4 ppm according to the fuel, so as not to be a source of problems.

TABLE 5-11. ESTIMATED SCR COST FOR NEW POWER PLANTS
(70% Boiler Utilization = 6,132 h/year.
Square honeycomb catalyst = 3.5 million yen/m³.
Power = 15 yen/kWh with coal, 18 yen/kWh with oil and gas.
Ammonia = 100,000 yen/t)

Fuel	Coal ^a	Oil ^a	Gas ^a	Coal ^b	Oil ^b	Gas ^b	Coal ^c	Oil ^c	Gas ^c
Capacity, MW	700	700	700	175	175	700	700	700	700
Flue gas, 1,000 Nm ³ /h	2,400	2,000	1,900	570	570	1,900	5,700	5,400	5,400
SCR inlet NO _x , ppm	500	200	120	60	1,000	200	1,000	200	100
80 _x , ppm ³	1,000	1,500	100	0	1,000	0	1,000	0	0
Dust, g/Nm ³	20	0.1	0.03	0.005	1	0	1	0	0
NO _x removal efficiency, %	80	80	80	80	80	80	80	80	80
SCR outlet NH ₃ , ppm	3	2	3	4	3	3	4	4	4
NH ₃ /NO _x mole ratio	0.82	0.84	0.83	0.86	0.81	0.95	0.86	0.86	0.92
Catalyst pitch, mm ³	7	6	6	5	7	7	7	5	5
volume, m ³	1,043	804	571	317	300	190	407	713	540
life, years	3	4	5	7	3	3	3	7	7
Space velocity, h ⁻¹	2,300	3,500	6,000	1,900	1,400	10,000	8,000	10,000	10,000
Pressure drop, mm H ₂ O	90	70	60	60	100	110	120	110	110
Power consumption, % ^e	0.22	0.20	0.16	0.14	0.23	0.25	0.60	0.52	0.52
Investment cost									
Catalyst, 10 ⁶ yen (A)	3,651	2,814	2,000	1,110	665	1,050	1,425	2,496	1,890
Other, 10 ⁶ yen (B)	1,217	1,400	1,200	1,000	600	650	2,200	2,200	2,200
Total, 10 ⁶ yen	5,151	4,214	3,300	2,310	1,765	1,765	4,096	4,096	4,096
Annual cost, 10 ⁶ yen	7,360	6,020	4,710	3,300	2,520	9,430	11,860	6,710	5,840
Fixed cost (0.1 A + 0.25 B)	740	631	525	411	342	255	306	800	739
Catalyst (A/life)	1,217	938	500	222	95	350	475	357	270
Power	141	128	124	108	108	38	39	463	401
MH	458	223	154	91	51	216	243	456	231
Other ^g	100	100	90	80	70	50	50	150	150
Total	2,656	2,020	1,393	912	666	909	1,103	2,226	1,791
Annualised cost									
Yen/kWh	0.62	0.47	0.32	0.21	0.16	0.84	1.02	0.52	0.42
Yen/Nm ³ of NO _x removed	449	731	709	816	1,184	324	350	398	677

^aConventional boiler (2 reactors)

^bWet bottom boiler (1 reactor)

^cCombined cycle (5 reactors)

^dDistance between center of walls

^ePercent of power generated

^fApproximate figure including ammonia system and civil engineering

^gApproximate figure

The calculated investment cost in Yen/kW for the conventional boilers is about 6,000-7,300 for coal-firing with inlet NO_x concentrations of 250-500 ppm, 3,300-4,700 for oil-fired boilers using low- and high-sulfur oils, and 2,500 for gas-firing. For a wet bottom, coal-fired boiler emitting a high concentration of NO_x , a large amount of catalyst is required and the investment cost is more than about 9,400 for 80% removal and almost 11,900 for 90% removal. The high cost is partly due to the relatively small size of the boiler and the SCR system.

For the combined cycle plant, each of the 5 trains is assumed to have a capacity of 140 MW with an SCR reactor. The investment cost is calculated to be about 5,800-6,700, which is much higher than for the conventional gas-fired boiler; the flue gas volume at the combined cycle plant is much greater and the NO_x concentrations are higher.

The annualized SCR cost in yen/kWh, including 7 years depreciation and 10% interest, is about 0.5-0.6 for coal-fired, 0.2-0.3 for oil-fired, and only 0.16 for gas-fired conventional boilers. The cost is about 0.8-1.0 for a wet bottom boiler with 80-90% removal and about 0.4-0.5 for the combined cycle. On the other hand, the annualized cost in yen/ Nm^3 of NO_x removed is the lowest with the wet bottom, coal-fired boiler and the highest with the gas-fired conventional boiler.

A gas-fired combined cycle plant may emit as much NO_x as a coal-fired, dry bottom boiler because the combined cycle gas volume is much larger while the NO_x concentration is lower. Compared with the SCR plant for coal, combined cycle SCR applications may require a nearly equal investment cost because of the larger gas volume, but have a lower annualized cost because of the longer catalyst life.

The catalyst requirement is estimated at a low unreacted NH_3 concentration of 2-4 ppm and may be much less if a greater amount of unreacted NH_3 is allowed. For example, with 10 ppm of unreacted NH_3 for a wet bottom coal-fired boiler, the catalyst requirement for 90% NO_x removal may be nearly

equivalent to that for 80% removal with 3 ppm unreacted NH_3 . Thus with the larger amount of unreacted NH_3 , the SCR cost can be substantially reduced but the air preheater may in turn accumulate ammonium bisulfate deposits and require water washing every few months. If the boiler is used for operation at peak load and can be shut down frequently, operation with less catalyst and a higher unreacted NH_3 concentration might be feasible.

Over the years, actual SCR cost has been lowered due to technological progress and further cost reductions may result from continued development of SCR technology. The current prices of the SCR catalyst and plant include a certain margin to allow the manufacturers to recover the cost of research and development. SCR prices will be considerably lower after the producers recover these expenses.

SECTION 6

VARIOUS DRY PROCESSES FOR NO_x AND SIMULTANEOUS SO_x/NO_x ABATEMENT

6.1 INTRODUCTION

Dry processes may be better suited than wet processes for NO_x and simultaneous SO_x/NO_x removal. There are several reasons for this. Wet NO_x and NO_x/SO_x removal processes produce wastewater containing nitrogen compounds which are not readily removed. Although it may be possible to produce a nitrogen fertilizer as a by-product from the wastewater, the process would be costly. Secondly, nearly all of NO_x in flue gases is in the form of NO, which is not readily absorbed in solutions. NO_2 formed by the oxidation of NO is more soluble but is still less soluble than SO_2 . Moreover, the oxidation reaction is expensive.

Between 1972 and 1978, approximately 30 small plants for wet process NO_x or simultaneous SO_x/NO_x removal were constructed. Nearly all of the plants that have been constructed more recently use dry processes. The dry processes other than SCR are shown in Table 6-1. Among the dry NO_x removal processes, selective noncatalytic reduction (SNR or Thermal De NO_x) is relatively advanced and has been applied to about 40 boilers and furnaces. In-furnace NO_x reduction (IFR or reburning) has now begun to be applied to several commercial boilers, while molecular sieve adsorption has been used at a nitric acid plant for tail gas treatment. Dry simultaneous removal processes are still in the development stage, except for the cupric oxide process which has been applied to an oil-fired boiler.

TABLE 6-1. TABULATION OF DRY PROCESSES FOR NO_x AND SO_x/NO_x ABATEMENT
(Other than SCR)

Process	Major Developers
NO_x removal only	
Selective noncatalytic reduction (SNR, or Thermal DeNO _x) (\$6.2)	Exxon, Mitsubishi Chemical, Mitsubishi Heavy Industries, Mitsubishi Kakoki
In-furnace NO _x reduction (\$6.3) (IFR, or Reburning)	Mitsubishi Heavy Industries, Hitachi Zosen
Molecular sieve adsorption (\$6.5.3)	Union Carbide-Nissan Chemical
Simultaneous SO_x/NO_x removal (\$6.4)	
Activated carbon adsorption	Utistica, Electric Power Federation, Electric Power Development Co. Sumitomo Heavy Industries Mitsui Mining-Bergbau Forschung
Electron beam radiation (\$6.5.1)	Ebara Corp.
Cupric oxide (\$6.5.2)	Shell
Fluidized bed combustion (\$6.7)	Electric Power Development Co. Kawasaki Heavy Industries Mitsubishi Heavy Industries

The following sections describe the major NO_x and SO_x/NO_x removal processes, and SO_x and NO_x abatement by fluidized bed combustion of coal. The respective section and subsection numbers are given in Table 6-1. Wet processes are described briefly in Section 6.6.

6.2 SELECTIVE NONCATALYTIC REDUCTION (SNR OR THERMAL DeNO_x)

6.2.1 Overview

Ammonia reacts with NO_x rapidly at 900-1,000°C without a catalyst to form N₂ and H₂O. Above 1,000°C, a considerable portion of the ammonia is oxidized to NO_x, while below 900°C the reaction occurs slowly, resulting in a low NO_x removal efficiency and a high concentration of unreacted ammonia.

In the U.S., Exxon has developed an NO_x removal technology called Thermal DeNO_x. In Japan, Mitsubishi Chemical, Mitsubishi Heavy Industries, Mitsubishi Kakoki, etc. have also developed the technology. In the Exxon technology, NO_x can be removed even at 700-800°C using hydrogen together with ammonia. Those technologies may be called selective non-catalytic reduction (SNR).

SNR was studied extensively in the mid-1970s when dirty gases were causing problems for SCR catalysts. The key to successful SNR is to mix ammonia rapidly and uniformly with the flue gas in the right temperature range. This can be done fairly easily in a small-scale plant but presents problems in a large-scale plant, particularly for utility boilers which undergo frequent temperature fluctuations due to load changes. Laboratory tests indicate that more than 90% of the NO_x can be removed using 2 mol NH₃ to 1 mol NO_x at 900-1,000°C. With commercial boilers, 2 mol NH₃ usually removes 50-60% of NO_x and produces 20-40 ppm of unreacted NH₃. The high unreacted NH₃ concentration may cause an ammonium bisulfate problem with the heat recovery system downstream when the flue gas contains SO_x (as was seen to be the case with SCR in Section 5).

Only a few SNR plants have been constructed for boilers since 1980 since SCR has been extensively developed during that time. However, more than 10 SNR units were recently constructed for city refuse incinerators for which 40-60% NO_x removal is required.

A combination of SNR and SCR has been tested by utility power companies (10). At the combination plant, a small amount of SCR catalyst was placed downstream from the SNR unit at 300-400°C, so that 50-70% of NO_x might be removed while maintaining unreacted NH_3 below 10 ppm. However, the tests were given up because of the complexity of the system and the thermal damage to the ammonia injection nozzle at temperatures of 900-1,000°C.

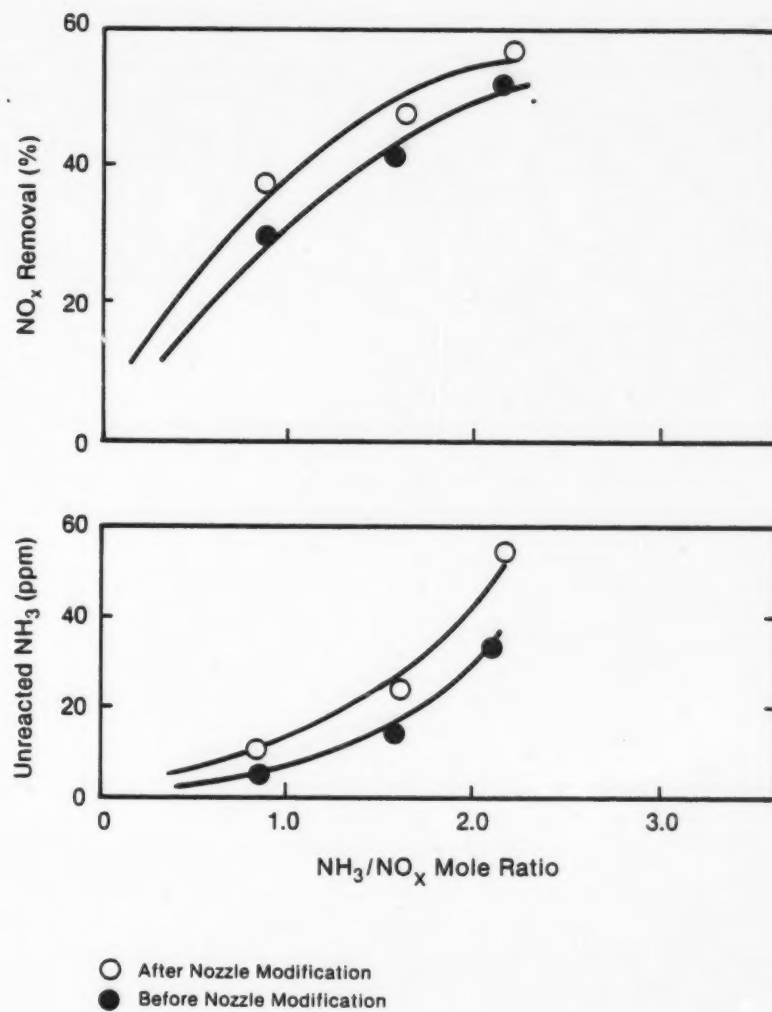
6.2.2 Chita Plant, Chubu Electric

MHI constructed a full-scale SNR unit for a 375 MW oil-fired boiler (0.2% S oil) at Chubu Electric's Chita Station. By using 1 mol NH_3 to 1 mol NO_x , 30-40% of NO_x has been removed while giving unreacted NH_3 concentrations of 10-15 ppm (Figure 6-1). After modification for greater removal effectiveness, 15 water-cooled nozzles with many holes were installed in two locations in the unit to cope with the temperature fluctuation (10). The investment cost was 600 million yen in 1976 (1,600 yen/kW).

6.2.3 Thermal De NO_x Units--Exxon Technology

Eight Thermal De NO_x units were constructed between 1975 and 1981 using Exxon technology (10). All of the units are for industrial boilers and furnaces with a capacity of 120,000-468,000 Nm^3/h , using mainly low-sulfur oil with some gas. Two units are for continuous use and remove about 20% of the NO_x using 0.4 mol NH_3 to 1 mol NO_x at 850-900°C. Five units are operated occasionally or during emergency periods of high photochemical smog and reduce 10-55% of the NO_x with 0.3-2 mol NH_3 to 1 mol NO_x together with H_2 at 700-1,000°C. The first unit, which was constructed in 1975, has now been shut down.

One of the plants is at Kawasaki and has a capacity of treating 254,000 Nm^3/h of flue gas from a pipestill furnace. An SCR catalyst has been installed to comply with the tightened regulations. Normally the plant is operated with SCR only to remove 80% of NO_x . However, it is expected that if required, about 90% of NO_x could be removed by the combination of Thermal De NO_x and SCR.



11-84-17714

Figure 6-1. Performance of SNR (Chita Station, Chubu Electric).

A new unit, scheduled to start operation in September 1984 at the Kawasaki works of Tonen Sekiyu Kagaku, will use an improved Thermal DeNO_x process to treat 195,000 Nm³/h of flue gas from an industrial boiler which is fired with low-sulfur oil and gas (25). This flue gas contains 170 ppm NO_x. In this new technology, ammonia is injected into the boiler at 850-1,050°C in a jet stream through 6 wall injectors placed in three locations on both side walls. About 30% of the NO_x will be removed by using 0.8-2.0 mol NH₃ to 1 mol NO_x. In contrast, in conventional SNR units, ammonia nozzles (grids) are inserted deep into the furnace where they are subjected to temperatures of 900-1,000°C and undergo thermal damage. The wall injector process requires a lower investment cost and less maintenance, and has been used by Exxon at demonstration units in the U.S. The Kawasaki unit may be the first commercial application of the injector system.

6.2.4 SNR Units by MKK

In 1975-1977, Mitsubishi Kakoki K.K. (MKK) constructed about 20 SCR units, mainly for the Toho Gas Co. The units have a flue gas treatment capacity ranging from 3,000 to 10,000 Nm³/h for low-sulfur oil or gas-fired applications. The NO_x removal efficiency ranges from 45 to 60%.

6.2.5 SNR for City Refuse Incinerators

MHI recently constructed many SNR plants for city refuse incinerators as shown in Table 6-2. Incinerator flue gas contains 150-200 ppm NO_x. Ammonia is injected into the incinerator at 700-1,100°C so that 40-60% of the NO_x is removed, leaving 60-120 ppm NO_x, as required by the local regulations.

Incinerator flue gas also contains various kinds of dusts with a considerable amount of HCl and is not easily treated by SCR. SNR may be more suitable to such a gas for NO_x removal levels of around 50%. Recently MHI has developed a catalyst useful for such applications. This catalyst will be used for an incinerator where an NO_x concentration below 50 ppm is needed.

TABLE 6-2. SNR UNITS FOR CITY REFUSE INCINERATORS

Location	Number of Units	Unit Capacity (Nm ³ /h)
Toyohashi	2	24,500
Yokohama	3	119,500
Kawasaki	3	43,500
Kobe	3	40,300
Kamakura	2	23,000

6.2.6 Evaluation

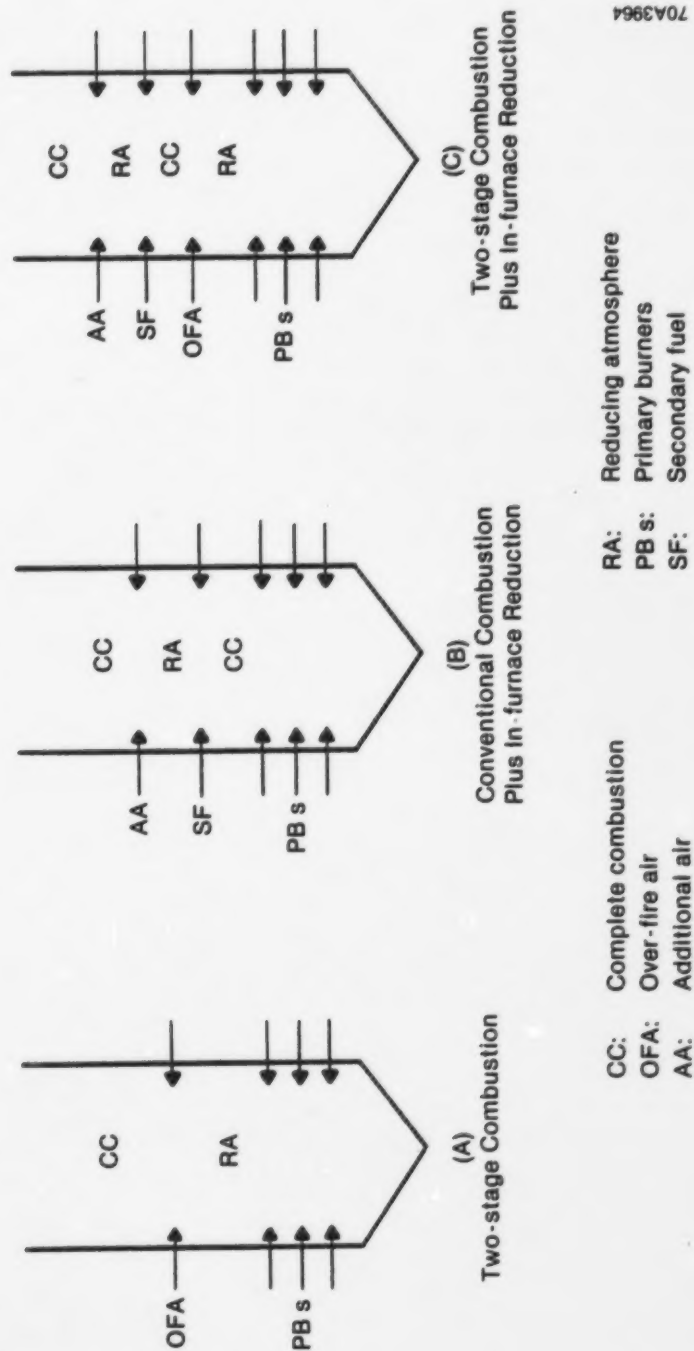
SNR produces a considerable amount of unreacted NH_3 and, therefore, should be used for gases containing little or no SO_3 , (to avoid the ammonium bisulfate problem), or for a relatively small amount of gas which can be emitted above 250°C. SNR may be especially suitable for certain types of furnaces with relatively long gas retention times at 900-1,000°C where there is a need to remove about 50% of the NO_x . It may also be particularly suitable to city refuse incinerator gas containing various kinds of dust and a considerable amount of chloride that may affect an SCR catalyst.

The new Exxon technology which uses wall injectors requires lower investment and maintenance costs but seems to consume a larger amount of ammonia than other types of SNR. Ammonia is much more costly in Japan than in the U.S. The technology may be particularly suitable to units used only for emergencies.

6.3 IN-FURNACE NO_x REDUCTION (IFR)

6.3.1 Overview

For IFR, about 10% of the fuel is injected into the boiler or furnace above the flame to reduce NO_x , as shown in Figure 6-2, items (B) and (C). IFR



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Figure 6-2. Two-stage combustion and in-furnace NO_x reduction.

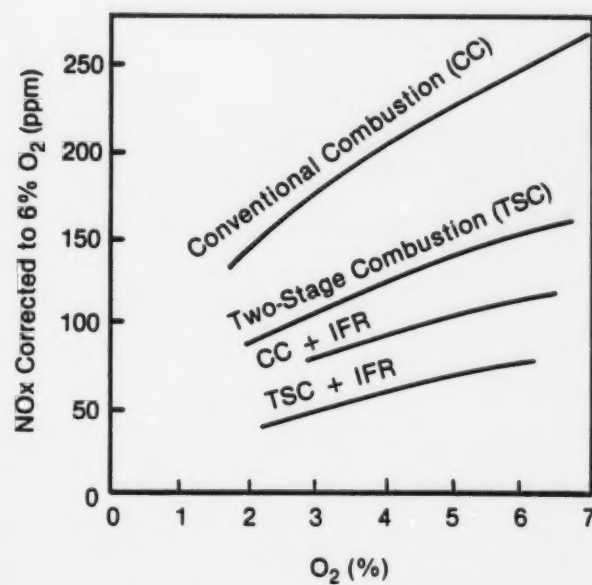
has also been developed in the U.S. where the process is called "reburning." In Japan, IFR has been developed by several organizations. Mitsubishi Heavy Industries has conducted extensive tests of an IFR process called "MACT," and has applied MACT to a few commercial oil-fired boilers. Hitachi Zosen calls the process "three stage combustion" and has conducted tests at pilot plants with flue gas from coal-firing. Tokyo Electric conducted extensive tests jointly with Mitsubishi Heavy Industries, Babcock Hitachi, and Ishikawajima-Harima Heavy Industries, but the test results have not been disclosed. Kobe Steel applied the IFR technology to substantially reduce NO_x from cement kilns.

The tests conducted by Mitsubishi Heavy Industries, Hitachi Zosen, and Kobe Steel are described in the following three subsections.

6.3.2 Mitsubishi Heavy Industries (MHI)

After conducting extensive testing, MHI has found that by injecting about 10% of the fuel above a $1,300^\circ\text{C}$ flame, nearly 90% of the NO_x can be removed. However, when air is added for more complete combustion, some NO_x forms, resulting in an overall NO_x removal efficiency rate of about 50% (10). Other test results with coal are shown in Figure 6-3. By combining IFR with two-stage combustion, NO_x production from the firing of domestic low nitrogen coal was less than 100 ppm.

Based on their studies, MHI applied IFR (MACT) to a few commercial oil-fired boilers. One of them is at the Yokkaichi Refinery of Mitsubishi Petrochemical. The boiler has an equivalent capacity of 80 MW and burns low-grade heavy oil that is rich in nitrogen and sulfur. The boiler uses two-stage combustion to lower NO_x to about 250 ppm, which is higher than the 150 ppm level required by local agreement. Therefore, IFR is used with addition of about 7% of the fuel oil as the secondary fuel, as shown in Figure 6-2, item (C). By this procedure, NO_x has been lowered to 130 ppm at full load and to about 100 ppm at reduced load. The operation has experienced no problems since startup in 1983. In order to apply IFR, the boiler was



11-84-17714

Figure 6-3. Test of IFR with domestic low-N coal (1,150 kg/h, 3 burners each from 4 corners).

constructed about 1 meter higher (about 5% of the total height) than normal. No appreciable increase in unburned carbon has been noticed.

6.3.3 Hitachi Zosen's IFR Tests

Hitachi Zosen has conducted tests of IFR (three stage combustion) at two pilot plants with capacities of 0.2 t/h and 2 t/h coal. The smaller pilot furnace is a horizontal one, while the larger one is vertical with an arrangement as shown in Figure 6-2, item (B). Test results with the larger plant burning imported coals--Austrian coal with 1.6% N and South African coal with 2.0% N--are shown in Figures 6-4 and 6-5. When the O_2 concentration in the flue gas was about 4%, the NO_x concentration from conventional combustion was about 700 ppm, but this was lowered by three-stage combustion to about 150 ppm (Figure 6-4). Figure 6-5 indicates that reducing the O_2 concentration in the flue gas below 4% further reduces NO_x , but considerably increases both unburned carbon in the fly ash and CO in the flue gas. Tests also indicated that the unburned carbon content of the ash was about 1% more with three-stage combustion than with conventional combustion.

6.3.4 Dual Combustion and Denitration Process for Cement Kilns (Nihon Cement-Kobe Steel Process)

Kobe Steel, jointly with Nihon Cement Co., has developed the Dual Combustion and Denitration Process (DD Process) to increase the production capacity and reduce NO_x emission concentrations. Following pilot plant tests, the process has been applied to commercial cement kilns since 1976.

The flowsheet and the specifications of the DD system as applied to the Kamiiso Plant of Nihon Cement Co., were shown in Figure 4-12 and Table 4-7 in Section 4.5 and a drawing of the DD furnace is shown in Figure 6-6. About half of the total fuel is injected into the DD furnace to lower the heat load in the rotary kiln and reduce NO_x formation. The DD furnace has a $DeNO_x$ burner for further NO_x abatement. A small amount of fuel is injected in Zone I so that NO_x formed in the rotary kiln is reduced. Using heavy oil firing

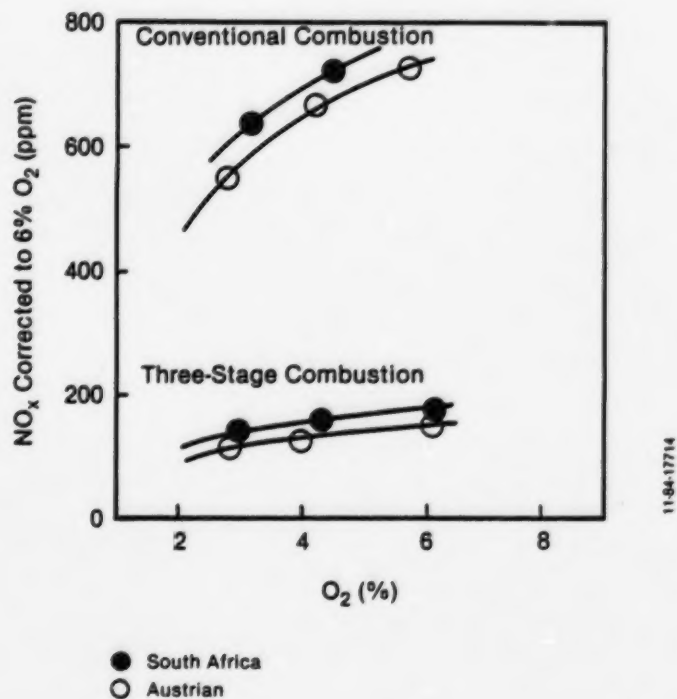


Figure 6-4. Tests with imported coals (opposite firing).

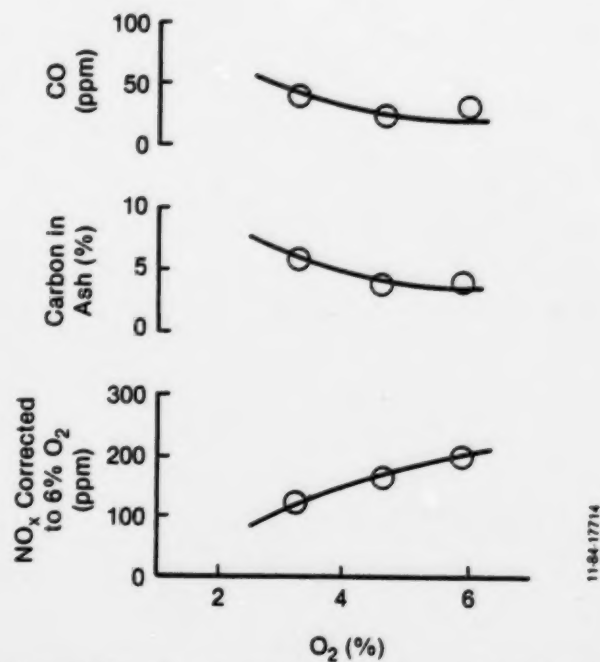
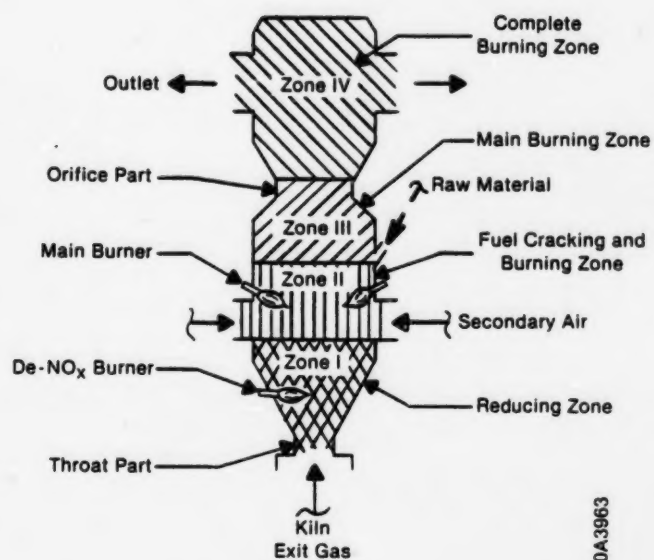


Figure 6-5. Tests with Austrian coal (front firing).



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Figure 6-6. Structure and function of DD furnace.

for the entire system, the NO_x concentration produced at 10% O_2 basis was about 400 ppm without using the DD furnace, 195 ppm using only the main burner of the DD furnace, and 110 ppm when the De NO_x burner was used. Recently coal has been used for the fuel. The NO_x concentrations have been kept below 400 ppm using the main burner and can be reduced to about 300 ppm using the De NO_x burner with the main burner.

6.3.5 Evaluation

IFR is simple, requires no special reducing agent such as ammonia, and experiences no ammonium bisulfate deposition problem. IFR's only disadvantage is that it requires a larger furnace volume: about 5% larger than for a furnace with two-stage combustion, and about 10% larger than that required for conventional combustion. Therefore, IFR may not be applied to most existing furnaces and boilers, but may be useful for new boilers and furnaces for NO_x abatement of about 50%.

Compared with selective noncatalytic reduction (SNR), which is also capable of 50% NO_x reduction, IFR is simpler and seems easier to operate because it is not subject to trouble with temperature fluctuation, other than slight changes in the NO_x removal rate. Conversely, with SCR, NH_3 may undergo partial conversion into NO_x when the temperature is too high, or may remain largely unreacted (excessive unreacted NH_3) when the temperature is too low.

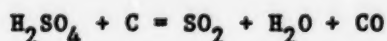
6.4 ACTIVATED CARBON (COKE) PROCESS

6.4.1 Introduction

Activated carbon (or activated coke) adsorbs SO_2 , O_2 , and H_2O from flue gas to form H_2SO_4 . The carbon works also as a catalyst for SCR NO_x removal. Catalyst activity can be promoted by adding a metal such as vanadium, but the addition increases the possibility of burning the carbon. NO_x reduction with ammonia occurs more rapidly at temperatures above 200°C, but SO_2 adsorption is

depressed at higher temperatures. Therefore, simultaneous SO_x/NO_x removal by carbon is usually conducted between 130 and 230°C.

In the activated carbon process, a partial reaction occurs between NH_3 and the H_2SO_4 contained in the carbon, forming NH_4HSO_4 . Washing the carbon with water yields a dilute solution of H_2SO_4 and NH_4HSO_4 which may have a very limited by-product use: e.g., a small amount may be used in fertilizer production. On the other hand, heating the carbon (containing H_2SO_4 and NH_4HSO_4) above 350°C in an inert gas or by indirect means, gives concentrated gaseous SO_2 which may be used for the production of elemental sulfur or sulfuric acid. The heating, however, not only requires a considerable amount of energy, but also causes the loss of the carbon due to the following oxidation reaction:



In 1975-1978, Unitika Co. conducted pilot plant tests of simultaneous SO_x/NO_x removal using the activated carbon process, but decided not to use the process because of the large amount of costly carbon consumed.

In West Germany, Bergbau Forschung developed the activated coke process for SO_2 and simultaneous SO_2/NO_x removal. In the U.S., Foster Wheeler developed the Resox process to convert concentrated SO_2 gas to sulfur using a coal reactor. Those two processes were combined as the BF-FW process for further development but the process has not been commercialized yet. One of the problems associated with the BF-FW process is the low sulfur recovery rate (about 60%) of the coal reactor.

In Japan, the Electric Power Development Co. (EPDC) jointly with Sumitomo Heavy Industries (SHI) has developed the activated coke process to produce elemental sulfur. EPDC uses the activated coke produced by Bergbau Forschung, which is less costly than the carbon used by Unitika.

Mitsui Mining Co., jointly with Mitsui Miike Machinery Co. and Bergbau Forschung, has also been trying to develop the activated coke process to by-produce elemental sulfur. In combination with this effort, Mitsui Mining operates a pilot plant for the production of activated coke. In addition, the Federation of Electric Power Companies has made similar tests at three pilot plants constructed by Mitsubishi Heavy Industries, Babcock-Hitachi, and IHI. However, the test results have not been disclosed. Therefore, the following two subsections will describe only the EPDC-SHI process and the Mitsui-BF process.

6.4.2 EPDC-SHI Process

Following tests at the Takehara pilot plant, which has a flue gas treatment capacity of $10,000 \text{ Nm}^3/\text{h}$ from a coal-fired boiler, a $300,000 \text{ Nm}^3/\text{h}$ demonstration plant attached to a coal-fired boiler at the EPDC Matsushima Station started operation in early 1984. A flowsheet of this process is shown in Figure 6-7; the process is basically the same as the one used at the Takehara pilot plant. The demonstration plant is designed to remove more than 95% of the SO_2 and 25% of the NO_x . However, the test results at the demonstration plant have not yet been disclosed.

At the Takehara pilot plant, the flue gas leaving an electrostatic precipitator at $120\text{--}150^\circ\text{C}$ was combined with 0.5 mol NH_3 to 1 mol SO_2 and passed through the activated coke in a moving bed. Over 90% of SO_2 was adsorbed by the carbon with an $\text{SV} = 500 - 900 \text{ h}^{-1}$ at $130\text{--}150^\circ\text{C}$ while 20-40% of NO_x was removed (Figure 6-8). The carbon was heated to temperatures above 350°C by a hot inert gas in a separate moving bed. The concentrated SO_2 that is released is then reduced to elemental sulfur by a coal reactor and Claus furnaces. The reduction process is called the "Reclaus process." By maintaining the $\text{SO}_2:(\text{H}_2\text{S} + \text{COS})$ mole ratio at 1:2, over 90% of the sulfur could be converted into elemental sulfur. The daily carbon consumption was 0.5-1% of the total process carbon.

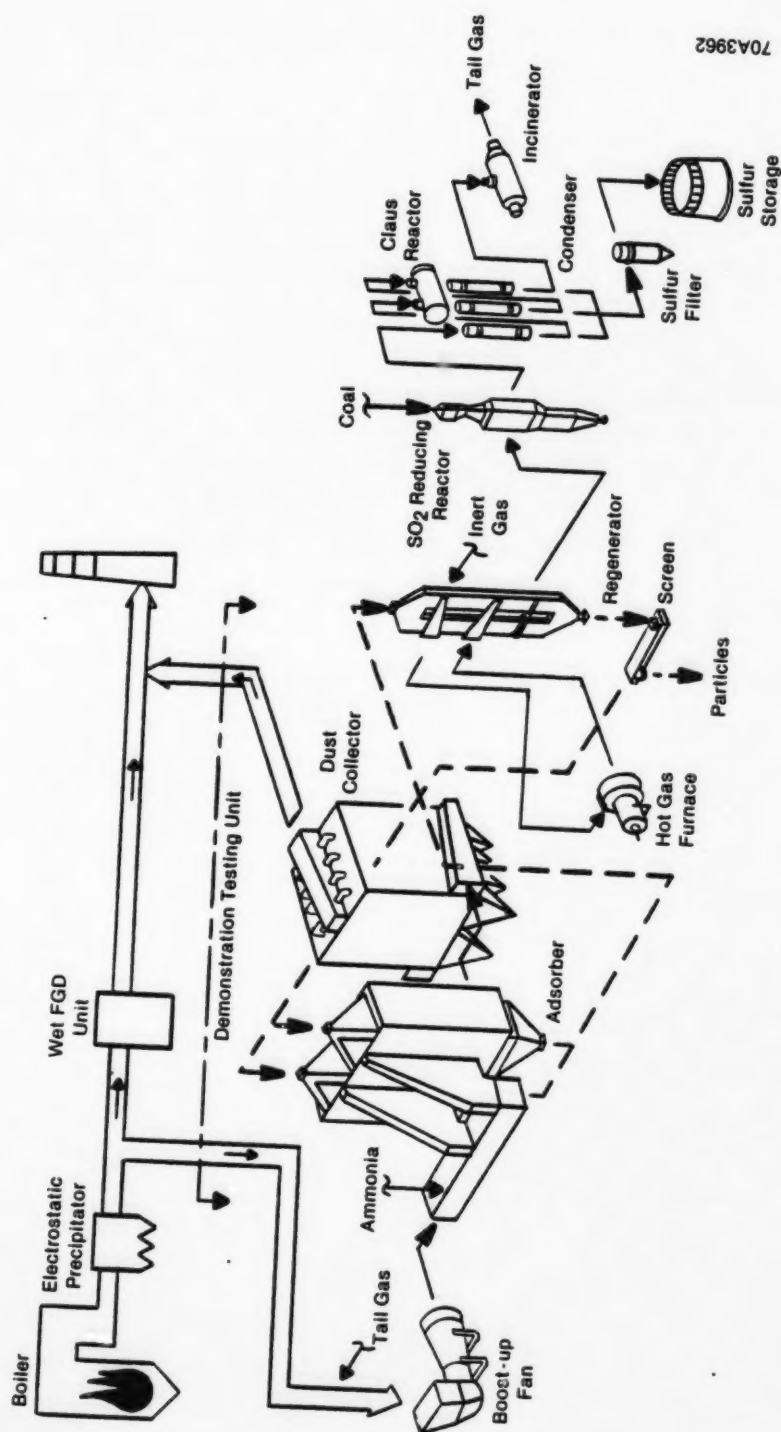
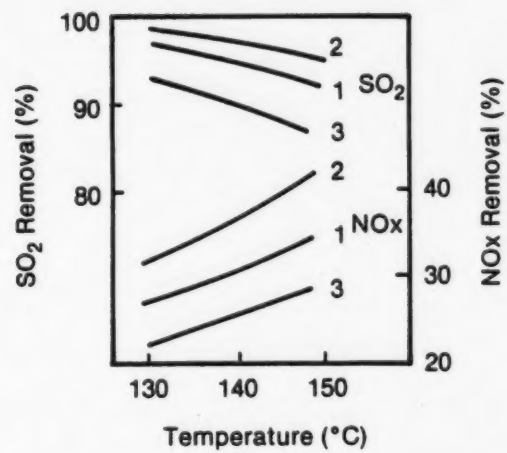


Figure 6-7. Flow sheet of EPDC, Matsushima Station demonstration unit for dry FGD with activated coke.



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	Relative Gas Volume	Inlet (ppm)	
		SO ₂	NOx
1	100	910	230
2	80	910	170
3	120	910	200

Figure 6-8. Test results of the activated carbon process at the Takehara pilot plant.

EPDC also has been constructing a 3,000 Nm³/h pilot plant at Matsushima to combine the activated carbon process with a two-stage reactor in which over 95% of the SO₂ and 80% of the NO_x can be removed. Since the process consumes a considerable amount of the carbon and might be more costly than the combination of wet process FGD and SCR, EPDC and Mitsui Mining Co. have started developing a production process for low cost activated coke.

6.4.3 Mitsui-BF Process

Mitsui Mining Co. has been developing an activated coke process for simultaneous SO_x/NO_x removal jointly with Mitsui Miike Engineering Co. (formerly MMMC) and Bergbau Forschung of West Germany. A pilot plant with a capacity of treating 1,000 Nm³/h of flue gas from an oil-fired boiler has been operated since 1981. A prototype plant with a 30,000 Nm³/h capacity for flue gas from a coal-fired boiler will be completed in the fall of 1984 at Omuta.

A flowsheet of the Mitsui-BF process is shown in Figure 6-9. Flue gas at 130°C enters a moving bed adsorber in which activated coke flows downward and the gas flows from one side to the other. There are two such beds in the adsorption vessel. The flue gas first flows through the lower one to remove most of the SO₂ with or without the addition of NH₃. The gas then is mixed with NH₃ and passed through the upper bed crossways to remove NO_x and most of the remaining SO₂.

Pilot plant tests with a flue gas containing 1,000 to 1,200 ppm SO₂ and 180 to 300 ppm NO_x showed that about 90% of the SO₂ was removed in the first stage, giving an overall SO₂ removal efficiency of over 98%. NO_x removal efficiency was 80% or higher. The coke leaving the adsorber is then subjected to indirect heating at 400°C to release the SO₂, which is treated by a coke reactor and Claus furnaces in a similar manner to the EPDC process. The pilot plant has been operated successfully. Activated coke consumption is about 2% per cycle, about half of which is due to oxidation and the rest to attrition.

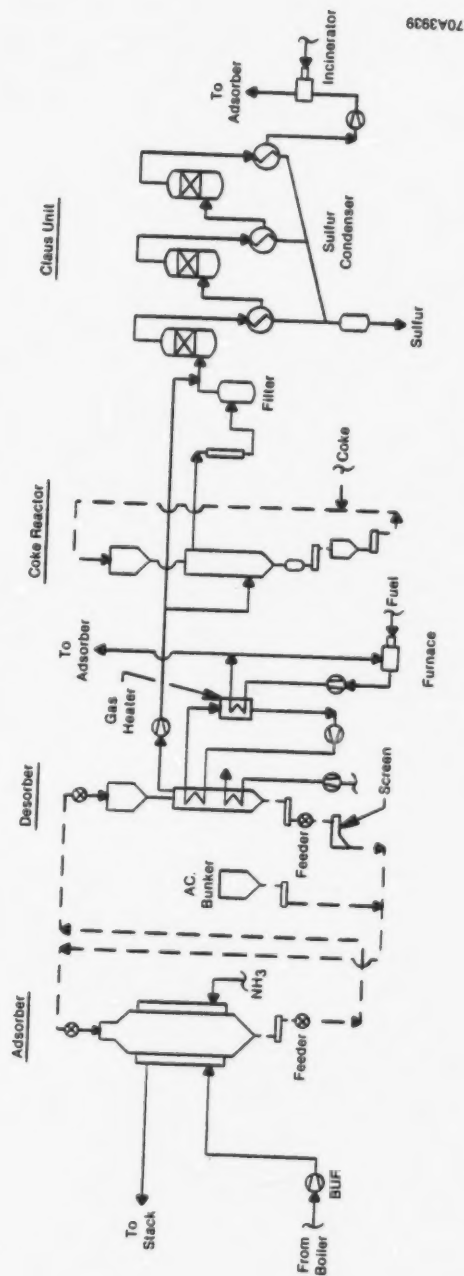


Figure 6-9. Mitsui Mining Process for removing NO_x and SO_2 by carbon adsorption.

At the prototype plant at Omuta, the flue gas will contain 500-1,000 ppm SO₂ and 150 ppm NO_x. The activated coke circulation rate will be 500-600 kg/h to remove over 98% of the SO₂ and 50-80% of the NO_x.

Table 6-3 shows the operating cost of a plant for a 350 MW coal-fired boiler, as estimated by Mitsui Mining Co. The total operating cost may be slightly higher than that for a combined wet limestone-gypsum process FGD/SCR system. However, Mitsui estimates that the investment cost of the Mitsui-BF process is considerably lower than that for the combined process so that the annualized operation cost, including depreciation and interest, may be less for the Mitsui-BF process than for the combined SCR/FGD system. Also, in an attempt to manufacture sufficiently activated, but less expensive coke for the process, Mitsui Mining is operating an activated coke production pilot plant with a capacity of 0.5 t/day.

TABLE 6-3. ESTIMATED HOURLY OPERATING COST OF MITSUI-BF ACTIVATED COKE PLANT FOR 350 MW COAL-FIRED BOILER (Yen)

Process Commodity	Rate		Cost (Yen)
Electric power	(15 yen/kWh)	15 x 2,800	= 42,000
Activated coke	(150 yen/kg)	150 x 697	= 105,000
Ammonia	(100 yen/kg)	100 x 310	= 31,000
Coke for reduction	(17 yen/kg)	17 x 2,000	= 34,000
Coal for heating	(20 yen/kg)	20 x 1,800	= 36,000
Claus catalyst	(2,500 yen/kg)	2,500 x 4	= 10,000
Water	(30 yen/t)	30 x 10	= 300
Total			258,000

6.4.4 Evaluation

The SO_2 and NO_x removal and the elemental sulfur recovery steps of the Mitsui-BF process are similar to the two-stage process of EPDC-SHI. The Japanese processes can attain a higher sulfur recovery efficiency than Foster Wheeler's Resox process (90% vs. 60% for Resox) but are more complex, requiring Claus furnaces. The activated coke process might be applied to oil refineries where there are existing Claus furnaces but seems too complex to be used at power plants.

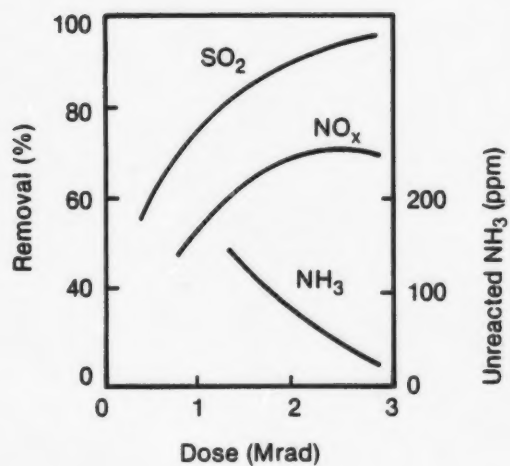
6.5 OTHER DRY PROCESSES

6.5.1 Electron Beam Process

Ebara Corporation and the Japan Atomic Energy Research Institute have developed a unique process to remove SO_x and NO_x simultaneously using electron beam radiation in the presence of ammonia. Under irradiation, SO_x and NO_x form fine crystals of ammonium sulfate and nitrate which are caught by an ESP or baghouse. The by-product is useful for fertilizer. In 1978, a pilot plant for this process with a flue gas treatment capacity of 3,000-10,000 Nm^3/hr was operated to remove 70-90% of the NO_x and SO_x produced by an iron ore sintering machine (Figure 6-10).

The process has been licensed to AVCO in the U.S., which conducted tests and found that fly ash from coal produced no adverse effect on the process. However, over 70% of the fly ash from pulverized coal should be removed prior to irradiation to make the product useful for fertilizer. AVCO estimates that the investment cost for this process is less than that for conventional wet lime scrubbing, provided that a large electron beam gun is used. Such a gun may be commercialized in the near future.

Ebara estimates that the power consumption for treating the flue gas from a coal-fired boiler with this process is about 3% of the power generated by the boiler. This percentage is slightly higher than that for the combination



Gas: 90°C, 3,000 Nm³/hr.,
 SO₂ 200 ppm,
 NO_x 200 ppm,
 NH₃ 600 ppm

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Figure 6-10. Test results of electron beam process.

of SCR (about 0.2%) and FGD (about 2.0%). A pilot plant for treating the flue gas from a coal-fired boiler (8 MW equivalent) is being sponsored by the U.S. Department of Energy and is currently under construction in the U.S.

6.5.2 Shell Copper Oxide Process

The Shell copper oxide process was applied to the Yokkaichi plant of SYS to treat the flue gas from an oil-fired boiler that contained about 1,000 ppm SO_x and 200 ppm NO_x . For several years, the plant was operated to remove only SO_x , and then was altered to remove approximately 40% of the NO_x and 90% of the SO_x , using ammonia. However, plant operation has been abandoned presumably because of the high operating cost. The boiler currently burns a low sulfur oil with combustion modification and without FGD. Only when further NO_x reduction is required temporarily by an emergency such as a photochemical smog warning, is the gas injected with ammonia and passed through the reactor to reduce greater amounts of NO_x .

6.5.3 Molecular Sieve Process

At the Toyama plant of Nissan Chemical, a molecular sieve adsorption unit has been operated since 1976 to remove about 90% of the NO_x in a tail gas from a nitric acid plant. After the adsorption, the molecular sieve is heated to desorb the NO_x , which is returned to the acid plant. The initial charge of molecular sieves is still in use.

6.6 WET PROCESSES FOR NO_x AND SIMULTANEOUS NO_x/SO_x REMOVAL

Although wet processes for FGD use are widespread, wet processes for NO_x removal have the following problems:

- o NO is not readily absorbed into solutions.
- o NO_2 is relatively soluble but still is less reactive with solutions than SO_2 .

- o Oxidation of NO to NO₂ is costly.
- o The nitrate and nitrite by-products have limited use. Although it is possible to produce a nitrate fertilizer, this requires the concentration of the dilute by-product solution which is costly.

In Japan, various attempts have been made to develop a process which simultaneously removes SO_x and NO_x by using the SO₂ to reduce the NO_x to N₂ or NH₃. There are two types of simultaneous removal processes--the oxidation/reduction process, shown in Table 6-4, and the complex absorption process, shown in Table 6-5. The oxidation/reduction processes use ClO₂ or O₃ as the oxidizing agent. These processes have been applied to several relatively small commercial plants. Ozone is too costly to be used on a large scale. ClO₂ is less costly, but the by-product contains chloride and creates a disposal problem. At most commercial plants which use the oxidation/reduction process jointly developed by Sumitomo Metal and Fujikasui Engineering, 99% of SO₂ and 90% of NO_x are removed. A new plant constructed in 1984 removes 98% of SO₂ with 35% of NO_x, using less oxidizing agent. In this process, a portion of the removed NO_x is converted to N₂ and the rest to nitrates. Process wastewater containing various compounds is treated with a much larger amount of wastewater from the factories.

The complex absorption process does not use oxidizing agents, but, instead, uses EDTA and ferrous ions which promote NO absorption by forming a complex. The process has not been applied commercially because of its complexity. There has been little recent progress with wet process development in Japan.

6.7 FLUIDIZED-BED COMBUSTION (FBC) OF COAL

6.7.1 Development Status

A pilot plant for FBC (6 MW equivalent) at atmospheric pressure was operated at the Wakamatsu Station of the Electric Power Development Co. Construction of a larger test plant (50 MW equivalent) will start in 1985.

TABLE 6-4. OXIDATION/REDUCTION PROCESSES AND PLANTS

Process Developer	Oxidizing Agent	Absorbent	By-Product	Capacity (Nm ³ /hr)	Start Up
Sumitomo-Fujikasui	ClO ₂	NaOH	NaNO ₃ , NaCl, Na ₂ SO ₄	62,000	1973
				39,000	1973
				100,000	1973
				39,000	1975
				80,000	1976
Sumitomo-Fujikasui	O ₃	NaOH	NaNO ₃ , Na ₂ SO ₄	12,000	1975
				18,000	1975
Sumitomo-Fujikasui	ClO ₂	Mg(OH) ₂	Mg(NO ₃) ₂ , MgCl ₂ , MgSO ₄	200,000	1984
IHI	O ₃	CaCO ₃	Gypsum, N ₂	5,000 ^a	
MHI	O ₃	CaCO ₃	Gypsum, NH ₃	2,000 ^a	
Chiyoda	O ₃	CaCO ₃	Gypsum, Ca(NO ₃) ₂	1,000 ^a	

^aPilot plant operated in 1970s.TABLE 6-5. COMPLEX ABSORPTION PROCESS (USING EDTA AND FeSO₄)

Process Developer	Absorbent	By-Product	Capacity (Nm ³ /hr)
Chisso Eng.	NH ₄ OH	(NH ₄) ₂ SO ₄	500 ^a
Asahi Chemical	NaOH, CaCO ₃	Gypsum, N ₂	600 ^a
Kureha Chemical	CH ₃ COONa, CaCO ₃	Gypsum, N ₂	5,000 ^a
Kureha Chemical	KOH	Sulfur, N ₂	2,000 ^a
MKK - HZ	KOH	Sulfur, N ₂	Bench scale ^b

^aTested in 1970s.^bStudied in the early 1980s (10).

Several commercial FBC plants are being operated for industrial use. Two of them are relatively large ones (19 MW and 38 MW) which use limestone as the bed material to remove SO_2 . Others are smaller, burn low grade coal and do not use limestone. This section describes NO_x and SO_2 removal at the larger FBC plants.

6.7.2 Wakamatsu Pilot Plant (EPDC) (26)

From 1981 to 1983, the Coal Mining Research Center, the Electric Power Development Co. (EPDC), Babcock Hitachi, and Kawasaki Heavy Industries (KHI) operated a pilot FBC plant at EPDC's Wakamatsu Station. The plant had a capacity of producing 20 t/h steam (6 MW equivalent). It had a main bed cell (MBC) and carbon burn-up cell (CBC) and burned about 2.5 t/h of coal with a particle size of under 7 mm. Coals shown in Table 6-6 were used for the tests.

TABLE 6-6. COALS USED FOR FBC TESTS AT WAKAMATSU PILOT PLANT

	B	C	D	E	F	G	I
Heat value (kcal/kg)	6515	6720	6666	6749	6920	6390	4170
Volatile matter (%)	29.3	37.9	27.1	29.6	30.8	23.4	22.4
Fixed carbon (%)	53.6	47.4	58.1	56.4	54.6	57.3	32.4
N (%)	1.38	1.42	1.47	0.71	1.43	1.46	0.64
Combustible S (%)	0.64	0.59	0.40	0.58	0.38	0.54	0.20
Total S (%)	0.97	0.76	0.42	0.67	0.40	0.77	0.33

Limestone made up of particles less than 3 mm in size was used as the bed material. The MBC has an over-fire air port. Two stage combustion was applied to reduce NO_x . Figure 6-11 shows the NO_x concentration from the combustion at 820°C with different air ratios in the MBC. NO_x was considerably reduced when two-stage combustion with a small air ratio was

used. The combustion efficiency in the MBC was about 90% but was lower when the air ratio in the main bed was smaller (Figure 6-12).

Figures 6-13 and 6-14 show SO_2 removal efficiency. About 90% of SO_2 was removed by using 5 mol CaO to 1 mol SO_2 . The efficiency was lower when the air ratio in the main bed was smaller, indicating that O_2 promotes the reaction of SO_2 with CaO . The optimum temperature for SO_2 removal was about 800°C.

EPDC plans to start construction of a larger test plant with a capacity of producing 160 t/h steam (50 MW equivalent) in October 1985 at the Wakamatsu Station.

6.7.3 Sakito FBC Plant (MHI-FCC process) (27)

Mitsubishi Heavy Industries, working with FCC in the U.S., has developed an atmospheric FBC process and constructed a plant to produce 58.8 t/h steam (17 MW equivalent). The plant built for the Sakito Salt Co. began commercial operation in February 1984. It has an MBC (main bed cell) and CBC (carbon burn-up cell) and burns a blend of a local coal and an imported coal containing 0.91% N and 1.14% S. Limestone, under 3 mm in size, is used as the bed material. The temperature of the MBC ranges from 850-900°C. The relationship between the Ca/S mole ratio and SO_2 removal efficiency is shown in Figure 6-15. The NO_x concentration has been maintained at about 200 ppm by low-excess air and two-stage combustion (Figure 6-16). Coal crushed to a particle size less than 15 mm is used and the combustion efficiency from the MBC and CBC is over 98%. Dust is removed by a pre-collector, multicyclone, and baghouse. Any products have been thrown away thus far.

6.7.4 Mitsui Aluminum Co.--Wakamatsu Plant

Mitsui Aluminum Co. constructed an FBC plant with a capacity of producing 120 t/h of steam (35 MW equivalent) at the Wakamatsu Works (28). The plant was constructed by KHI, based on the experiences of the pilot plant operation

at EPDC's Wakamatsu plant nearby. The FBC plant has an MBC and CBC. By agreement with a local authority, NO_x must be kept below 120 ppm (4% O_2 basis), SO_x below 160 ppm (4% O_2 basis), and particulates below 50 mg/Nm³. A blend of Australian coal and domestic high-sulfur coal is used in an 80:20 ratio. The plant began test operation in October 1983 and commercial operation in May 1984. Operational parameters are shown in Table 6-7.

TABLE 6-7. DESIGN AND FBC OPERATIONAL DATA OF MITSUI ALUMINUM WAKAMATSU PLANT

	Design	Actual
Steam generation capacity, t/h	120	120-123
Steam pressure at superheater outlet, kg/cm ² g	63	62.6-63.8
Steam temperature at superheater outlet, °C	460	454-456
Feed water temperature, °C	175	173-175
Waste gas temperature at economizer outlet, °C	135	157-164
Boiler efficiency, percent	Over 90	Over 90
Bed load, kcal/m ² h MBC	1.2×10^6	1.2×10^6
- CBC	1.0×10^6	1.0×10^6
Standard operating temperature, °C - MBC	825	808-816
- CBC	850	825-830

Coal crushed to a particle size of less than 15 mm is used. The blended coal contains 1.5% nitrogen and 1.1% sulfur. Limestone particles between 1 and 3 mm are used. The plant applies two-stage combustion to reduce NO_x . NO_x levels have been kept at 84-89 ppm (4% O_2 basis) by maintaining the air ratio at the first stage (main bed) at about 1.0, and the SO_2 concentration has been kept at 96-116 ppm (at 4% O_2 basis), using 4.5 mol CaO to 1 mol SO_2 .

The ash from the MBC caught by the multicyclone is granulated by water addition and fed to the CBC. The overall combustion efficiency is 98%. The

by-product is rich in lime and thus is used to treat red mud, the by-product of alumina production from bauxite.

6.7.5 Evaluation

FBC may be useful for certain industrial boilers which are subject to moderate SO_x and NO_x regulations, particularly when the lime-rich, ash by-product has some use. FBC, however, may not be suitable for utility boilers in Japan because the process: 1) is not conveniently applied when there are frequent boiler load changes or startup and shutdown cycles; 2) is subject to considerable heat loss during shutdown because of the large heat content of the bed material; 3) is not easily used to meet stringent SO_x and NO_x regulations; and 4) creates a disposal problem because of the large amount of lime-rich ash by-product.

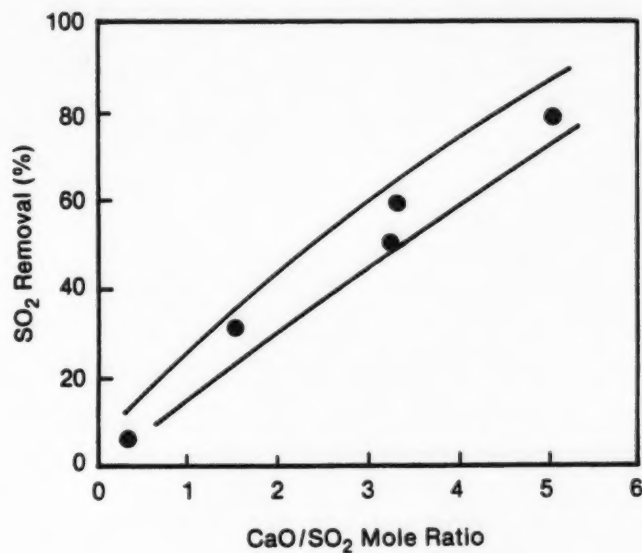


Figure 6-15. SO₂ removal efficiency at the Sakito FBC plant (850°-900°C).

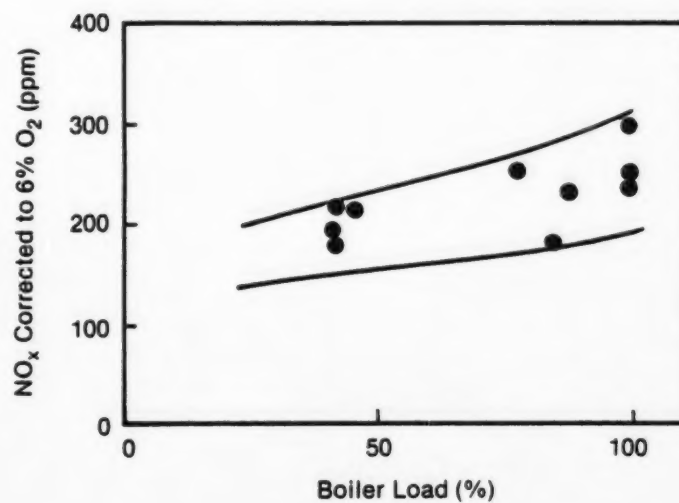


Figure 6-16. NO_x concentrations at the Sakito FBC plant (850-900°C).

TABLE 6-7. DESIGN AND FBC OPERATIONAL DATA OF MITSUI ALUMINUM
WAKAMATSU PLANT

	Design	Actual
Steam generation capacity, t/h	120	120-123
Steam pressure at superheater outlet, kg/cm ² g	63	62.6-63.8
Steam temperature at superheater outlet, °C	460	454-456
Feed water temperature, °C	175	173-175
Waste gas temperature at economizer outlet, °C	135	157-164
Boiler efficiency, percent	Over 90	Over 90
Bed load, kcal/m ² ·h MBC	1.2×10^6	1.2×10^6
- CBC	1.0×10^6	1.0×10^6
Standard operating temperature, °C - MBC	825	808-816
- CBC	850	825-830

Coal crushed to a particle size of less than 15 mm is used. The blended coal contains 1.5% nitrogen and 1.1% sulfur. Limestone particles between 1 and 3 mm are used. The plant applies two-stage combustion to reduce NO_x. NO_x levels have been kept at 84-89 ppm (4% O₂ basis) by maintaining the air ratio at the first stage (main bed) at about 1.0, and the SO₂ concentration has been kept at 96-116 ppm (at 4% O₂ basis), using 4.5 mol CaO to 1 mol SO₂.

The ash from the MBC caught by the multicyclone is granulated by water addition and fed to the CBC. The overall combustion efficiency is 98%. The by-product is rich in lime and thus is used to treat red mud, the by-product of alumina production from bauxite.

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SECTION 7

ESTIMATED COSTS FOR POLLUTION CONTROL AND EVALUATION OF CONTROL METHODS

7.1 POWER GENERATION AND POLLUTION CONTROL COSTS

Costs of power generation by coal- and oil-fired boilers, including pollution control, are calculated based on the assumptions shown in Table 7-1.

TABLE 7-1. COAL- AND OIL-FIRED BOILER POWER GENERATION, ASSUMPTIONS FOR COST ESTIMATION: NEW UTILITY BOILERS. (Two units each of 1,000 MW. SO₂ emissions = 100 ppm, NO_x emissions = 100 ppm, particulate emissions below 30 mg/Nm³)

	Sulfur (%)	Cost	Gas Cleaning Method
Coal	1.2	14,000 yen/ton	SCR, ESP and FGD, as shown in Figure 1-7.
Oil	0.2	55,000 yen/kl	Combustion modification and ESP

The calculated costs are shown in Table 7-2. The investment cost for coal-firing is much higher than for oil-firing but the annualized cost of the former is lower, even though about 20% of the total cost goes for pollution control.

TABLE 7-2 COST OF POWER GENERATION AND POLLUTION CONTROL (1984)

	Investment Cost (yen/kW)		Annualized Cost (yen/kWh) ^a	
	Coal	Oil	Coal	Oil
Power generation	210,000	170,000	12.2 ^b	17.4 ^c
FGD	20,000	-	1.4	-
SCR	6,000	-	0.5	-
ESP	4,000	3,000	0.1	0.05
Other pollution control ^d	20,000	7,000	1.0	0.15
Total	260,000	180,000	15.2	17.6

^aIncluding 7 years' depreciation for pollution control facilities

^bIncluding 6.4 for fuel

^cIncluding 12.9 for fuel

^dAsh disposal, water and noise control, etc.

The cost of oil-firing was higher in late 1972, when low-sulfur oil cost more than 60,000 yen/kl while coal cost more than 15,000 yen/ton. The estimated power generation cost at that time was about 21 yen/kWh with oil and 18 yen/kWh with coal (3). However, the price of oil decreased in 1983 so that the power generation cost was about equal for oil and coal, causing a delay in the construction schedule for some new coal-fired boilers. More recently, the price of coal and pollution control costs decreased so that once again coal has become more economical than oil, as shown in Table 7-2. The use of high-sulfur oil together with FGD and partial SCR may be of about equal or slightly greater cost than the use of low-sulfur oil.

Current power generation costs in yen/kWh for efficient existing power stations may be about 14 for coal, 17 for oil or natural gas (LNG), and 12.5 for nuclear power. It appears possible that nuclear energy costs will increase because of the requirement for further safety measures. If so, coal will be the least expensive energy source for power generation, even in Japan which has to import coal and requires stricter pollution control measures than other countries.

7.2 ACID REMOVAL COSTS

SO_2 and NO_x removal are important for prevention of acid precipitation. Table 7-3 compares acid removal costs by wet FGD, combustion modification (CM), and SCR. For the removal of acid (H^+), CM is the most economical, while SCR is more costly than FGD. For acid rain prevention, primarily CM and FGD should be used. SCR should be used in regions where NO_x is present in high concentrations and causes photochemical smog and adverse effects on health. Table 7-3 also indicates that the combination of SCR and CM may be less costly than SCR for attaining a high removal efficiency.

TABLE 7-3. ACID REMOVAL COSTS (Coal-fired Boilers)

Pollutant	SO_2		NO_x					
	FGD		CM		SCR			CM(1) CM(2)
Control technology			(1)	(2)	(1)	(2)	(3)	SCR(2) SCR(3)
Concentration, ppm								
Not treated	1500	600	600	600	600	400	250	600 600
Treated	100	50	400	250	100	100	100	100 100
Removal, %	93	92	33	58	83	75	60	83 83
Removal cost								
Yen/kWh	1.5	1.4	0.03 ^a	0.1 ^a	0.7	0.5	0.4	0.53 0.50
Yen/mole	7	17	1	2	9	14	18	6.6 6.3
Yen/mole H^+	3.5	8.5	1	2	9	14	18	6.6 6.3

^aRough figure which includes the loss of fuel from increases in the level of unburned carbon.

7.3 EVALUATION OF CONTROL METHODS

Both FGD and SCR have become highly reliable and less costly. New FGD plants for coal-fired boilers in Japan are usually operated by 2 persons per shift and remove approximately 95% of the SO_2 and 70-80% of the dust without problems. FGD plants are usually operated continuously for 11 months and then undergo annual maintenance for about a month during the boiler shut-down period.

The FGD plant cost has been lowered from 30,000-40,000 yen/kW in 1979-1982 to about 20,000 yen/kW. The annualized FGD cost, including 7 years depreciation and 10% interest, has been reduced from 2 to 1.4 yen/kWh. Further cost reductions may not be easily possible as long as the high SO_2 removal level of 95% is needed. FGD costs account for about 10% of the total power generation cost with coal and may be considered too high in many countries. Development of more cost-effective FGD processes is desirable before widespread application of FGD can be expected in many countries.

SCR technology has become even more reliable. SCR plants for utility boilers have been operated automatically and are experiencing no appreciable difficulties. SCR cost has been also considerably lowered and is currently about one-third that of FGD. Power generation by coal with SCR, ESP, and FGD applied is considerably less costly than with oil (refer to Table 7-1). Further SCR cost reductions can be expected from possible decreases in catalyst cost.

However, even with the cost reductions, SCR is considerably more costly than combustion modification (CM), as was shown in Table 7-2. By the application of advanced CM technology, including a low- NO_x burner, NO_x concentrations in the flue gas from coal-, oil-, and gas-fired utility boilers may be lowered to 150-250 ppm, 80-130 ppm, and 40-50 ppm, respectively. NO_x concentrations may be further reduced to about one-half by the use of in-furnace NO_x reduction (IFR). However, CM, including two-stage combustion, may require about 5% larger furnace volume than usual and IFR may need

yet another 5%. Still, CM is the best method for NO_x abatement, and IFR may be useful especially for new boilers and furnaces. SCR and other NO_x removal processes should be applied if further NO_x abatement is necessary.

Selective noncatalytic reduction (SNR or thermal DeNO_x) may be useful for relatively small boilers and furnaces which use low-sulfur fuels and require NO_x removal to levels below 50%. It may be suitable to a furnace with a relatively long gas retention time at 900-1,000°C, and also to special furnaces which yield a flue gas rich in impurities which might affect an SCR catalyst. The improved thermal DeNO_x process, using a wall injector, may be particularly suitable for emergency boiler use because of the low investment and maintenance costs and the possible high ammonia consumption.

Both SCR and SNR use ammonia and decompose it to N_2 and H_2O . If they are applied extensively in many countries in the future, the world ammonia supply may decrease, affecting fertilizer production and thus the food supply. NO_x removal processes that do not consume ammonia might therefore be more desirable as future technologies.

Simultaneous SO_x and NO_x removal by activated coke also consumes ammonia. The ammonia requirement to remove 1 mol of NO_x is about 1 mol with SCR, 2 mols with the activated coke process, and 3 mols with SNR. The electron beam process uses ammonia and yields ammonium sulfate and nitrate by-products useful for fertilizer. The process, however, seems costly, requiring electron beam accelerators and a considerable amount of electric power.

Wet simultaneous processes can convert NO_x into NH_3 or N_2 through reduction with SO_2 , but so far are too complex for commercial application. Wet processes can be simplified by the use of ozone but this may be too costly for large-scale commercial use. Drastic improvements or entirely new systems may be required for a wet process to be used on a large scale. Dry simultaneous removal processes also have difficulties, however, and require considerable improvement before commercial application is feasible.

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